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Quantum Experiments with Spatial Modes of Photons in Large Real and Hilbert spaces

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Abstract

Photons, the quanta of light, can occupy very complex spatial structures when they propagate freely. These structures are described by spatial modes, yielding an infinite-dimensional, discrete Hilbert space. An interesting property of photons in these light structures is that they can carry a quantized unit of angular momentum which is - at least theoretically - unbounded. This unrestricted state space allows, in principle, information coding with a large alphabet which enables information densities of more than one bit per photon.

This property is, of course, interesting in classical communication, where it is already used to achieve data rates of more than 100 Tbit/sec. Especially in quantum-physical situations, it also allows very interesting possibilities such as high-dimensional entanglement. Two systems are entangled if they jointly take one of several settings (such as the polarization of photons), but this setting is not realized before the measurement. Only by measuring one of the photons, the setting is realized (for example, that the photon has horizontal polarization). As it is a common setting of the photon pair, the second photon is then also defined in this setting – no matter how far they are apart. High-dimensional entanglement means that the photons can jointly take one of many settings. By measuring one of the photons, one of these settings is realized, and because of entanglement, this setting is also realized at the partner photon. This shows that two photons can carry a lot of information jointly – over long distances – in a non-local / non-realistic way.

In this work, I have dealt with spatial structures of photons and their quantum-physical properties - in large Hilbert spaces as well as in large real spaces.

In the first experiment, we investigate in how many dimensions two photons can be entangled. We were able to produce a pairs of photons, which are entangled in over 100 dimensions. The entanglement has been confirmed in a series of a total of 200,000 measurements. The corresponding Hilbert space is comparable to two-dimensional entangled system with 13 particles. This shows a huge potential for the implementation of quantum communication experiments with existing technologies.

Based on this, we asked ourselves whether photons can be transmitted in such light structures over distances that are larger than typical optical tables in the laboratory – which is of course necessary for realistic quantum communication experiments. This question was studied up to the time only in theoretical modeling and numerical "experiments", with quite critical results. However, since there were no experimental investigations, not even for classical lasers with spatial structures, we conducted a first test over a 3 kilometer long communication link through the city of Vienna. This first test showed (for us unexpectedly) good results. Motivated by that, in a follow-up experiment we investigated the transmission of entangled photons in

spatial structures over same 3 kilometer link. We could actually verify the property of entanglement, which means that the spatial structure of single photons is not influenced too much by the atmosphere. In order to exploit the limits further, we carried out a classic experiment between two Canary islands over 143 kilometers, and the transferred light structures could still be recognized.

In another work we wanted to extend the potential of high-dimensional entanglement to a multi-party system in order to investigate new effects in high-dimensional multi-particle entanglement. At that time, we encountered the following practical problem: We did not find an experimental setup which can produce such states. The problem I have solved by a computer algorithm (which we call MELVIN) that can automatically find implementations of experiments. Several of these computer-designed experiments have already been successfully implemented in our laboratories, others are being built at the moment. These resulting experiments allow the investigation of quantum states and properties in the laboratory, which can otherwise only be analyzed on paper. The computer-designed experiments are very counterintuitive and difficult to understand – but they also involve techniques that we would not normally think of. Such a technical *trick* has inspired us to discover a new kind of very general method to create entanglement. For me, this is an example of successful human-machine synergy.

In the last chapter, I have listed nine follow-up questions and projects, whose solution I would find particularly exciting.

Vorwort

Photonen, die Quanten des Lichts, können sehr komplexe räumliche Strukturen einnehmen, wenn sie frei propagieren. Diese Strukturen werden durch räumliche Moden beschrieben, und ergeben einen unendlich-dimensionalen, diskreten Hilbert-Raum. Eine interessante Eigenschaft von Photonen in diesen Lichtstrukturen ist, dass sie eine quantisierte Einheit von Drehimpuls tragen können, welcher – zumindest theoretisch – nach oben hin unbeschränkt ist. Dieser unbeschränkte Zustandsraum lässt im Prinzip Informationskodierung mit einem großen Alphabet zu, das Informationsdichten von mehr als einem Bit pro Photon ermöglicht.

Diese Eigenschaft ist natürlich in der klassischen Kommunikation interessant, wo sie bereits benutzt wird, um Datenraten von mehr als 100 Tbit/sec zu erreichen. Speziell in quantenphysikalischen Situationen lässt es auch sehr interessante Möglichkeiten zu, wie zum Beispiel hochdimensionale Verschränkung. Zwei Systeme sind verschränkt, wenn sie gemeinsam eine von mehreren Eigenschaften annehmen können (wie zum Beispiel die Polarisation von Photonen), diese Eigenschaft aber vor der Messung noch nicht realisiert ist. Erst durch die Messung von einem der Systeme wird eine Eigenschaft realisiert (zum Beispiel, dass das Photon horizontale Polarisation hat), und weil es eine gemeinsame Eigenschaft des Photonen-Paars ist, nimmt auch das zweite Photon diese Eigenschaft an – egal wie weit es entfernt ist. Hochdimensionale Verschränkung bedeutet, dass die Photonen eine von vielen Eigenschaften gemeinsame einnehmen könnten. Aber erst durch die Messung wird eine dieser Einstellungen realisiert – und im gleichen Augenblick nimmt das Partner-Photon diese Eigenschaft an. Das zeigt, dass zwei Photonen gemeinsam über große Distanzen hinweg auf eine nicht-lokale/nicht-reale Weise viel Information tragen können.

In dieser Arbeit habe ich mich mit räumlichen Strukturen von Photonen und ihren quantenphysikalischen Eigenschaften beschäftigt – in großen Hilbert-Räumen als auch in großen realen Räumen.

Im ersten Experiment wird untersucht, in wie vielen Dimensionen man zwei Photonen verschränken kann. Wir konnten Photonenpaare erzeugen, die in über 100 Dimensionen verschränkt sind – und haben diese in einer Serie von insgesamt 200.000 Messungen auch nachweisen können. Der entsprechende Hilbert Raum ist vergleichbar mit zwei-dimensionaler Verschränkung von etwa 13 Teilchen. Das zeigt ein riesiges Potential für die Umsetzung von Quantenkommunikations-Experimenten mit bereits vorhandenen Technologien.

Aufbauend darauf haben wir uns gefragt, ob Photonen in diesen Lichtstrukturen auch über Distanzen übertragen werden können, die größer sind als typische optische Tische im Labor – was natürlich für realistische Quantenkommunikations-Experimente notwendig ist. Diese Frage wurde bis zu dem Zeitpunkt nur in theoretischen Modellierungen und numerischen "Exper-

imenten” untersucht – mit durchaus kritischen Resultaten. Da es aber noch keine experimentellen Untersuchungen gab, noch nicht einmal für klassische Laser mit räumlichen Strukturen, haben wir über einen 3 Kilometer langen Kommunikationslink – durch die Stadt Wien – einen ersten Test gemacht. Dieser erste Test zeigte (für uns unerwartet) gute Resultate, so dass wir in einem Nachfolgeexperiment auf dem gleichen 3 Kilometer Link auch verschränkte Photonen in räumlichen Strukturen übertragen haben. Wir konnten die Eigenschaft der Verschränkung tatsächlich nachweisen, was bedeutet dass die räumliche Struktur von Einzelphotonen durch die Atmosphäre nicht zu stark beeinflusst wird. Um die Limits weiter auszuschöpfen, haben wir ein klassisches Experiment zwischen zwei kanarischen Inseln über 143 Kilometer durchgeführt, und die übertragenen Strukturen konnten immer noch erkannt werden.

In einer weiteren Arbeit wollten wir das Potential von hochdimensionaler Verschränkung auf mehrere Teilchen erweitern, um neue Effekte in hoch-dimensionaler Vielteilchen-Verschränkung zu untersuchen. Dabei stießen wir gleich auf das folgende praktische Problem: Uns war nicht klar, wie der dementsprechenden experimentelle Aufbau aussehen sollte, der solche Zustände erzeugen kann. Das Problem habe ich gelöst durch einen Computeralgorithmus (den wir MELVIN nennen) der automatisch Implementationen von Experimenten finden kann. Mehrere dieser computer-designten Experimente wurden in unseren Labors bereits erfolgreich umgesetzt, andere werden im Moment gebaut. Diese daraus resultierenden Experimente erlauben die Untersuchung von Zuständen im Labor, die man sonst nur auf Papier analysieren kann. Die computer-designten Experimente sind sehr kontraintuitiv, und schwierig zu verstehen – sie beinhalten aber auch Techniken an die wir normalerweise nicht denken würden. Ein solcher technische *Trick* hat uns zur Entdeckung einer neuen Art von sehr allgemeiner Verschränkungserzeugung inspiriert, und ist (für mich) ein Beispiel von erfolgreicher Mensch-Maschine Synergie.

Im letzten Kapitel habe ich neun Folgefragen und -Projekte aufgelistet, deren Lösung ich besonders spannend finden würde.

Contents

Abstract	3
Vorwort	5
1 Introduction	9
1.1 Motivation	9
1.2 Overview	10
2 Background	12
2.1 From Maxwell to Paraxial Wave Optics	12
2.2 Transverse Spatial Modes	14
2.2.1 Gaussian mode	14
2.2.2 Laguerre-Gauss modes	14
2.2.3 Hermite-Gauss modes	16
2.2.4 Ince-Gauss beams	16
2.3 On an apparent paradoxon	18
2.3.1 Non-paraxial treatment of Laguerre-Gauss modes	19
2.3.2 Applications to small Laguerre-Gauss modes	21
2.3.3 Remarks	24
2.4 Quantum States and Mutually Unbiased Bases	25
2.5 Two-Party Quantum States and Quantum Entanglement	26
2.5.1 Bell basis	29
2.5.2 High-dimensional entanglement	29
3 Quantum Entanglement in 100 dimensions	31
3.1 Motivation	31
3.2 High-dimensional entanglement witness	32
3.2.1 Two-dimensional subspace entanglement witness	33
3.3 Experimental implementation	35
3.3.1 Considered Modes vs. Entanglement Dimensionality	40
3.3.2 Effect of Deviation from Perfect Correlation	41
3.4 Discussion	42
4 Long-distance transmission of spatial modes	43
4.1 Motivation	43
4.2 Classical transmission over an intra-city link	44
4.2.1 Experimental setup	45
4.2.2 Mode detection	48

Contents

4.2.3	Data transmission encoded in OAM	49
4.2.4	Relative phase stability	50
4.2.5	Remarks	51
4.3	Quantum Entanglement over an intra-city link	52
4.3.1	Experimental setup	53
4.3.2	Measurement results	56
4.3.3	Remarks	64
4.4	Classical transmission over 143 kilometers	65
4.4.1	Experimental setup	66
4.4.2	Transmission quality	68
4.4.3	Data transmission	71
4.5	Discussion	72
5	Automated Search for new Quantum Experiments	74
5.1	Motivation	74
5.2	The algorithm	75
5.2.1	Symbolic description language	76
5.2.2	Experimental components	77
5.2.3	Simplifying experiments	80
5.3	Example 1: High-dimensional multipartite entanglement	80
5.3.1	Details for the 3-dimensional GHZ-state experiment	83
5.4	Example 2: High-dimensional cyclic operations and learning	85
5.4.1	Learning from previous Experimental Designs	86
5.5	Experimental implementations	87
5.6	Example 3: Entanglement by Path Identity	90
5.6.1	Inspired by a computer algorithm	90
5.6.2	Path identity	91
5.6.3	Multi-photon entanglement in Polarisation	92
5.6.4	Realistic experimental diagram and coherence time requirements	93
5.6.5	Multi-photon entanglement in higher dimensions	94
5.6.6	Two-photon arbitrary high-dimensional entanglement	96
5.6.7	Construction of general experiments	97
5.7	Discussion	99
6	Conclusion and Outlook	100
7	Bibliography	105
	Appendix	120
	Experimental setups for other Cyclic Operations	120
	List of Publications	122
	Acknowledgements	124
	Curriculum Vitae	126

1 Introduction

1.1 Motivation

Since ancient times, light is a window into the universe. Not only does it allow to observe the cosmos ¹, but the systematic investigation into the properties of light has also revealed many secrets of the world on the small scale.

In more modern days, the experimental study of light has lead to the first observations of quantum mechanical phenomena (*Fraunhofer lines* as discrete dark lines in the spectrum of the sun in the early 19th century; the *black-body radiation* in the 1850-1860s by Robert Kirchhoff and others, the *photoelectric effect* which was observed by Heinrich Hertz in the 1880s, to name a few), albeit they have not been understood at that time.

When considering that light has been studied since this long time, it is all the more surprising that new features are still being identified. Twenty-five years ago, in 1992, it has been found that a certain type of transverse spatial modes is connected with an orbital angular momentum (OAM) [1]. Transverse spatial modes are properties of freely propagating light beams, which do not have a Gaussian shape. These transverse spatial profiles can be decomposed into an infinitely large, discrete set of modes. One of these mode families – the Laguerre-Gauss modes – possess this special property. In 1995, it has been directly observed experimentally that a beam with well-defined OAM can rotate macroscopic objects (small black ceramic particles with a diameter in the order of $1\mu\text{m}$) [2]. A few years later, it was shown that two photons can be entangled in that property [3], showing that individual photons can have a well-defined orbital angular momentum quantum number. These initial investigations have lead to a large interest in the topic of spatial modes of photons, ranging from fundamental studies of topological structures, classical communication, trapping of particles, microscopy, and investigations of quantum mechanical properties, to name a few [4–6].

Since spatial modes have a large, discrete Hilbert space, and their quantum number correspond, in addition, to a physical degree of freedom, they can be studied in manifold ways. In this manuscript I summaries my investigations into spatial modes of photons in *extremal* situations. In particular, I investigated the following questions:

- How do strongly focused beams with very large angular momentum behave?
- How can high-dimensional two-photon entanglement be created and confirmed?
- How can high-dimensional multi-photon entanglement be created?

¹Starting from cave paintings in Lascaux of celestial objects from 15.000 years ago, to the first (known) star catalogue by Claudius Ptolemaeus 2.000 years ago.

1 Introduction

- How can spatial modes of light be transmitted over large distances?
- How can photonic entanglement encoded in spatial modes be transmitted over large distances?

Many of the answers have raised more general follow-up questions (which I elaborate on in the last chapter).

1.2 Overview

The thesis is structured in the following way:

In **Chapter 2** it is shown how the paraxial wave equation – whose solutions are used in the following chapters of this thesis – is obtained from Maxwell’s equation. One instructive example is given showing an effect due to the breakdown of the paraxial approximation. Afterwards, I briefly introduce quantum states, mutually unbiased bases and quantum entanglement.

In **Chapter 3**, I describe an experiment in which we observed 100-dimensional quantum entanglement between two photons. The entanglement was encoded in the transverse spatial modes of photons, using both the angular and radial quantum number. For that, an experimentally feasible entanglement dimensionality witness (Schmidt number witness) has been implemented, which requires measurements of two-dimensional subspaces only. The experiment indicates the potential of spatially entangled photon pairs for increasing the capacity in quantum communication protocols.

Chapter 4 contains descriptions of three experiments in which we investigate long-distance transmissions of (quantum) information encoded in the spatial modes. Those modes can not be transmitted through fibers, and until recently it was believed that those modes do not survive free-space propagation over a distance of more than 1 kilometer, due to influence of atmospheric turbulence. In our first experiment, we show that classical information can be encoded in spatial modes, transmitted over a 3-kilometer-link in Vienna, and decoded afterwards. Motivated by these results, we conducted two follow-up experiments. In one of them, we distribute quantum entanglement over the same 3 kilometer link. The entanglement was encoded (in the transmitted photon) in spatial modes. The result shows that both single-photon spatial coherence as well as two-photon coherence is not significantly deteriorated by the turbulent link. In a different follow-up experiment, we extended the distance for classical communication from 3 kilometers in Vienna to 143 kilometers between two Canary islands. All of these experiments show that classical and quantum information can be transmitted through turbulent free-space links, and the application of already existing adaptive optics (such as tip-tilt mirrors) could significantly improve the transmission qualities.

In **Chapter 5**, I explain a computer algorithm which we use to find new implementations of quantum experiments which can be realized in our labs. The motivation came from a situation when we wanted to create a multi-photon multi-dimensional entangled quantum state, but were not able to find an experimental implementation ourselves. The program found a feasible

1 Introduction

experimental proposal. It also found solutions to other questions which we were not able to solve by hand. The program can also learn from previous solutions, and thereby reducing significantly the time to discover more complex solutions. The final examples shows that the program can not only be used to find solutions to well-posed questions, but can also be used as an inspiration for human scientist. Several of the computer-designed experiments have been implemented successfully in our laboratories, others are being built at the moment.

In the final **Chapter 6**, I conclude and elaborate nine follow-up questions and projects which I would find genuinely exciting.

2 Background

2.1 From Maxwell to Paraxial Wave Optics

In the first section, we will start from Maxwell's equations – the equations describing the behaviour of electromagnetic fields, and finally arrive at the paraxial wave equation – whose solutions are used in later chapters. For this, we mainly follow Saleh & Teich [7], Siegman [8] and Lax, Louisell & McKnight [9]. The propagation of light is described by Maxwell's equations¹. In the absence of charges and currents ($\rho = 0$ and $J_i = 0$), they are written as

$$\text{div} \mathbf{E} = \nabla \cdot \mathbf{E} = 0, \quad (2.1)$$

$$\text{div} \mathbf{B} = \nabla \cdot \mathbf{B} = 0, \quad (2.2)$$

$$\text{curl} \mathbf{E} = \nabla \times \mathbf{E} = -\frac{\partial}{\partial t} \mathbf{B}, \quad (2.3)$$

$$\text{curl} \mathbf{B} = \nabla \times \mathbf{B} = \mu_0 \epsilon_0 \frac{\partial}{\partial t} \mathbf{E}. \quad (2.4)$$

where $\mathbf{E}$ and $\mathbf{B}$ are the electric and magnetic fields respectively, ϵ_0 and μ_0 are the vacuum permittivity and permeability (with speed of light in vacuum $c = \frac{1}{\sqrt{\epsilon_0 \mu_0}}$). We apply the *curl* at both sides of equation (2.3), which leads to

$$\begin{aligned} \nabla \times \nabla \times \mathbf{E} &= -\nabla \times \frac{\partial}{\partial t} \mathbf{B} \\ (\text{grad div} - \Delta) \mathbf{E} &= -\frac{\partial}{\partial t} \nabla \times \mathbf{B}. \end{aligned}$$

Using equation (2.1) and (2.4), we find the **electromagnetic wave equation**

$$\left(\Delta - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) \mathbf{E}(\mathbf{r}, t) = 0. \quad (2.5)$$

For the magnetic field, an analog derivation can be applied. For monochromatic waves, the time-dependence reduces to $\mathbf{E}(\mathbf{r}, t) = \mathbf{E}(\mathbf{r}) \cdot \exp(i\omega t)$ where ω is the wavelength. This leads to the **vectorial Helmholtz equation**

$$(\Delta + k^2) \mathbf{E}(\mathbf{r}) = 0, \quad (2.6)$$

where $k = \frac{\omega}{c}$ is the wave number. For a plane wave propagating in the z-direction, we can write $\mathbf{E}_{pw}(\mathbf{r}) = \mathbf{E}_0 \cdot \exp(ikz)$, and from Maxwell's equation (2.1) we find

$$\text{div} \mathbf{E}_{pw} = k \cdot E_z = 0 \quad (2.7)$$

¹Maxwell's original equations published in 1865 consisted 20 equations and 20 unknowns [10], which have been simplified to the modern version by Oliver Heaviside [11].

2 Background

which means that the electric field is transversal, i.e. $\mathbf{E}(\mathbf{r}) = (E_x(\mathbf{r}), E_y(\mathbf{r}), 0)$. This condition holds exactly only for plane waves, but it is a good approximation for propagating beams when the beam waist is much larger than the wave length (which will be discussed more in chapter 2.3).

The transversality condition (2.7) shows that the electric field components into the direction of propagation vanishes. Here, an electric field with vanishing E_y component is called horizontally (H) polarized, and with vanishing E_x component is denoted as vertically (V) polarized. Fields with both E_x and E_y components can be written in the form

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_H(\mathbf{r}) + a \cdot \mathbf{E}_V(\mathbf{r}) \quad (2.8)$$

If $a = 1$ or $a = -1$, the beam is diagonal (D) or antidiagonal (A) polarized. If $a = i$ or $a = -i$, the two components have a phase shift and are called right (R) or left (L) circular polarized. In the scalar wave approximation, the wave equation reduces to the **scalar Helmholtz equation** (which can be done when the vectorial character of the field can be neglected, such as in the simple case of linear polarized fields $E_y(\mathbf{r}) = 0$ and $E_x(\mathbf{r}) = U(\mathbf{r})$):

$$(\Delta + k^2)U(\mathbf{r}) = 0. \quad (2.9)$$

For the construction of paraxial waves, one can use the ansatz of a plane wave with a slowly varying envelope in the z-direction,

$$U(\mathbf{r}) = A(\mathbf{r}) \cdot \exp(-ikz). \quad (2.10)$$

The paraxial approximation assumes *slowly varying* fields in z-direction. This means, within the distance of $\Delta z = \lambda$, the beam should vary only slowly $|\Delta A| \ll |A|$ [7]. That leads to

$$\frac{\partial A}{\partial z} \ll kA \quad (2.11)$$

$$\frac{\partial^2 A}{\partial z^2} \ll k \frac{\partial A}{\partial z} \quad (2.12)$$

Substituting (2.10) into (2.9) and taking into account the paraxial approximation (2.12), one finally arrives² at the **paraxial wave equation**

$$\left(\Delta_T - 2ik \frac{\partial}{\partial z} \right) A(\mathbf{r}) = 0, \quad (2.13)$$

where $\Delta_T = \partial_x^2 + \partial_y^2$. The approximation breaks down for strongly focused beams, where the vectorial character of the electromagnetic field comes into play. A detailed investigation into the scope of the paraxial wave equation has been done by Lax, Louisell and McKnight in 1975 [9], and in simpler way by Davis in 1979 [12]. They found that the paraxial wave equation appears as the zero order in an expansion of the Maxwell's equation in powers of $(\frac{\lambda}{w_0})$, where w_0 is the beam waist and λ is the wavelength. That means, as long as the beam waist is much larger than the wavelength ($w_0 \gg \lambda$), the paraxial wave equation can describe the situation sufficiently well.

Throughout this manuscript I will use solutions of the paraxial wave equation, with one exception in chapter 2.3 where we investigate one peculiar case of spatial modes in a strongly focused regime.

²Using $U(\mathbf{r})'' = (A(\mathbf{r})'' + 2ikA(\mathbf{r})' - k^2A(\mathbf{r})) \exp(-ikz)$

2.2 Transverse Spatial Modes

First we explain briefly the Gaussian mode, and from there investigate higher-order modes.

2.2.1 Gaussian mode

The simplest solution of the paraxial wave equation (2.13) is the Gaussian mode $A(\mathbf{r}) = \psi_G(\mathbf{r})$. It can be written as [8] (a detailed and instructive derivation can be found in [13])

$$\psi_G(\mathbf{r}) = N \cdot \frac{w_0}{w(z)} \cdot \exp\left(\frac{r^2}{w(z)^2} - i\left(k\frac{r^2}{2R(z)} - \phi_G(z)\right)\right) \quad (2.14)$$

where N is a normalisation constant, w_0 is the beam waist in the focus,

$$w(z) = w_0 \sqrt{1 + \left(\frac{z}{z_R}\right)^2} \quad (2.15)$$

is the radius where the field amplitude falls to $\frac{1}{e}$; and $z_R = \frac{\pi w_0^2}{\lambda}$ is the Rayleigh range.

$$R(z) = z \left(1 + \left(\frac{z_R}{z}\right)^2\right) \quad (2.16)$$

is the radius of curvature and finally

$$\phi_G(z) = \arctan\left(\frac{z}{z_R}\right) \quad (2.17)$$

is the Gouy phase, which accounts for the change of the phase while the mode passes through the focus.

By solving the paraxial wave equation using separation of variables, higher-order transverse spatial modes can be found. Different orthogonal coordinate systems lead to a different complete and orthogonal set of basis modes, such as the **Laguerre-Gauss modes** in cylindrical coordinates, the **Hermite-Gauss modes** in cartesian coordinates or the **Ince-Gauss modes** in elliptical coordinate systems.

2.2.2 Laguerre-Gauss modes

Laguerre-Gauss (LG) modes can be derived by separating the variables of the paraxial wave equation in cylindrical coordinates

$$\begin{pmatrix} x \\ y \end{pmatrix} = \begin{pmatrix} r \cdot \cos \phi \\ r \cdot \sin \phi \end{pmatrix}. \quad (2.18)$$

It leads to

$$LG_{n,\ell}(r, \phi, z) = \underbrace{\psi_G(\mathbf{r})}_{\text{Gaussian envelope}} \overbrace{\left(\frac{\sqrt{2}r}{w(z)}\right)^{|\ell|} L_n^{|\ell|}\left(\frac{2r^2}{w(z)^2}\right)}^{\text{radial modulation}} \underbrace{\exp(i\ell\phi)}_{\text{spiral phase}} \overbrace{\exp\left(-i(|\ell| + 2n)\phi_G(z)\right)}^{\text{mode dependent Gouy phase}} \quad (2.19)$$

Here, $n \in \mathbb{N}_0$ and $\ell \in \mathbb{Z}$ are two independent mode numbers, called radial and angular momentum mode number. $L_n^{|\ell|}$ stands for the Laguerre polynomials, with the property $L_0^{|\ell|}(x) = 1$ (i.e.

2 Background

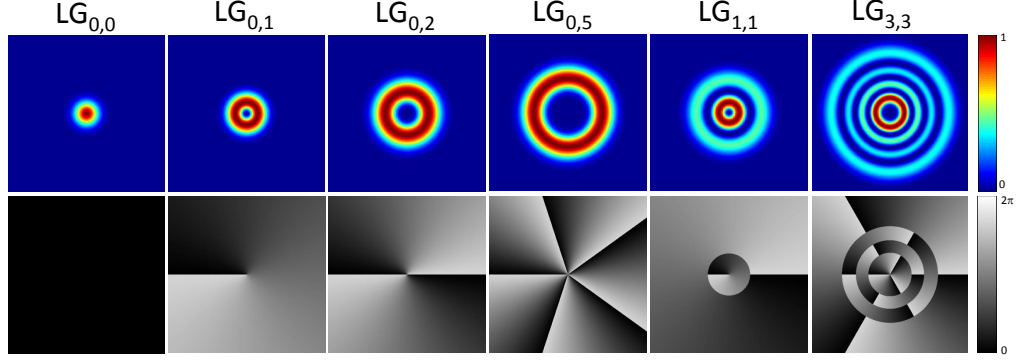


Figure 2.1: *Intensity and Phase of Laguerre-Gauss modes with different mode number.* The upper row shows the intensity distribution, the lower row shows the transverse phase distribution. $LG_{0,0}$ stands for Laguerre-Gauss mode with both mode numbers being zero – which is the Gauss mode. Phase jumps in radial direction are in steps of π . However, the apparent phase jumps in azimuthal direction are in steps of 2π , which is not observable (because a phase $\phi = 2\pi = 0$) (Figure from [14]).

for $n = 0$, the radial modulation is governed by $\psi_G(\mathbf{r})r^{|\ell|}$. For $|\ell| > 0$, the mode has an intensity-zero in the center at $r=0$ which leads to ring-shaped intensity structures. Furthermore, it has a spiral (or *twisted*) phase structure. Within one rotation around the center, it changes its phase from zero to $2\pi\ell$. This phase corresponds to an orbital angular momentum of the light [1]. The physical property has been experimentally shown first in [2] in 1995, by transferring angular momentum to μm -scale reflective particles. The mode has a phase singularity at $r = 0$ as the phase is undefined at that position, and ℓ is sometimes denoted as the strength or topological charge of the singularity or vortex [15].

The second quantum number, n , stands for the additional rings of the mode in radial direction. Beside that, its physical interpretation is less clear as for the ℓ mode number. There is a differential operator for $\hat{N}$ where eigenstates are the LG modes and with eigenvalues of n [16–18]. It can be interpreted as a radial momentum (however due to $r \geq 0$, in a different coordinate system [18]).

The last term in (2.19) is a mode-dependent Gouy-like phase. Mode superpositions with the same mode order $p = (|\ell| + 2n)$ are propagation invariant as they collect the same Gouy phase. However, superpositions of modes with different $p = (|\ell| + 2n)$ change their shape while passing through the focus point, because each mode has a different z -dependent phase. The intensity and phase structure of LG modes with different mode numbers can be seen in figure (2.5).

One important feature is that the LG modes form a complete and orthogonal basis. Every paraxial beam of light can be uniquely decomposed into components of LG modes.

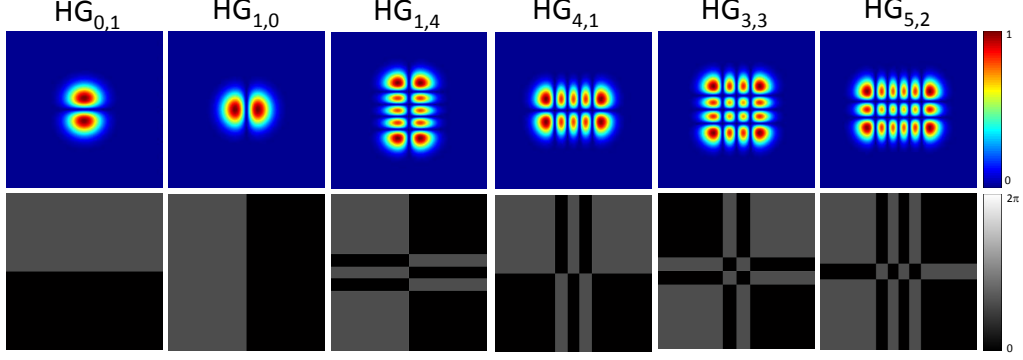


Figure 2.2: *Intensity and Phase of Hermite-Gauss modes with different mode numbers.* The upper row shows the intensity distribution, the lower row shows the transverse phase distribution. The mode numbers of $HG_{m,n}$ modes both correspond to the number of π -phase jumps in the x or y direction, respectively. The π -phase jumps lead to $m + 1$ or $n + 1$ intensity maxima in x -direction or y -direction.

2.2.3 Hermite-Gauss modes

Hermite-Gauss (HG) modes are best derived in cartesian coordinates, which leads to a solution that is symmetric in x and y [7]:

$$HG_{m,n}(r, \phi, z) = \underbrace{\psi_G(\mathbf{r})}_{\text{Gaussian envelope}} \overbrace{H_m\left(\frac{\sqrt{2}x}{w(z)}\right) H_n\left(\frac{\sqrt{2}y}{w(z)}\right)}^{\text{spatial modulation}} \underbrace{\exp\left(-i(m+n)\phi_G(z)\right)}_{\text{mode dependent Gouy phase}} \quad (2.20)$$

H_n and H_m are Hermite polynomials with mode numbers n and m both $\in \mathbb{N}_0$, which are responsible for the spatial modulation of the mode. The last term corresponds again to a mode-dependent Gouy-phase, which restricts the number of mode superpositions which lead to shape-invariant modes (again, as in the case for LG modes, only superpositions of modes with the same mode order $p = (m + n)$ propagate without changing their spatial structure). In contrast to LG modes, HG modes do not possess phase vortices but only phase jumps. In figure (2.20), the intensity and phase structure of these modes at $z = 0$ are illustrated.

As in the case of LG modes, HG modes also form a complete, orthogonal set - and each other paraxial beam can be decomposed into the HG basis. In particular, a Laguerre-Gauss beam can be decomposed into HG modes. One interesting property follows directly from the mode-dependent Gouy phase, which needs to be the same because of shape-invariance. As an example, a LG beam $LG_{n=1}^{\ell=0}$ and $LG_{n=0}^{\ell=2}$ have the same mode order $p = 2$, therefore their decomposition in the Hermite-Gauss basis can only contain modes with $p = 2$, which are $HG_{2,0}$, $HG_{1,1}$ and $HG_{0,2}$.

2.2.4 Ince-Gauss beams

Ince-Gauss beams are solutions of the paraxial wave equation in elliptical coordinates, which have been derived by Bandres and Gutiérrez-Vega in 2004 [19, 20], and observed for the first

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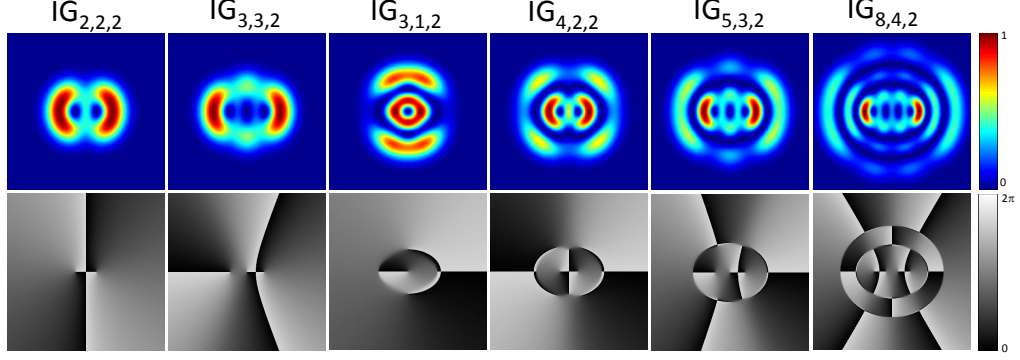


Figure 2.3: *Intensity and Phase of Ince-Gauss modes $IG_{p,m,\epsilon}$ with different mode number, with constant ellipticity $\epsilon = 2$. The upper row shows the intensity distribution, the lower row shows the transverse phase distribution. It can be observed that all higher-charge vortices (with charge m) split into unity-charge vortices. In addition, new singularities and vortices occur in the outer regions of the modes for cases when $\tilde{n} = \frac{p-m}{2} > 0$, where $\tilde{n}$ is the number of additional elliptical rings of the mode (for ellipticity $\epsilon \rightarrow 0$: $\tilde{n} \rightarrow n$, where n is the radial mode number of Laguerre-Gauss modes). (Figure from [14])*

time in a symmetry-broken laser cavity [21] and with optical holograms [22]. Elliptic coordinate can be written as

$$\begin{pmatrix} x \\ y \end{pmatrix} = f_0 \begin{pmatrix} \cosh u \cdot \cos v \\ \sinh u \cdot \sin v \end{pmatrix}. \quad (2.21)$$

The strength of the ellipticity is given by f_0 , which is the semi-focal separation (eccentricity), u is the radial elliptic coordinate, and v is the angular elliptic coordinate. The paraxial wave equation in the elliptical coordinates leads to the Ince equation, first studied by Edward Lindsay Ince in the 1920s [23]. Its solutions are given by

$$IG_{p,m,\epsilon}(u, v, z) = \underbrace{\psi_G(\mathbf{r})}_{\text{Gaussian envelope}} \underbrace{\overbrace{C_p^m(iu, \epsilon) C_p^m(v, \epsilon)}^{\text{spatial modulation}}}_{\text{mode dependent Gouy phase}} \exp\left(-ip\phi_G(z)\right)$$

where $C(x, \epsilon)$ stand for the Ince polynomials [24], and $\epsilon = \frac{2f_0^2}{w_0^2}$ is the ellipticity.

The beams are a generalisation of Laguerre-Gauss and Hermite-Gauss modes, which are a special case for $\epsilon \rightarrow 0$ and $\epsilon \rightarrow \infty$. For every constant value of ϵ , the modes form a complete orthogonal basis set. The intensity and phase structure of several modes with constant ϵ can be seen in figure (2.3). For a finite ellipticity ϵ , multiple charged vortices split and additional phase vortices can appear in the phase structure, as one can observe in figure (2.4).

While Laguerre-Gauss beams carry an integer number of $\hbar$ per photon of angular momentum, for non-vanishing ellipticity, the Ince-Gauss modes are not eigenstates of the angular momentum operator, but have a complicated, ellipticity-dependent average value of angular momentum per photon [26]. Also, entanglement between photons in such modes has been investigated recently [27].

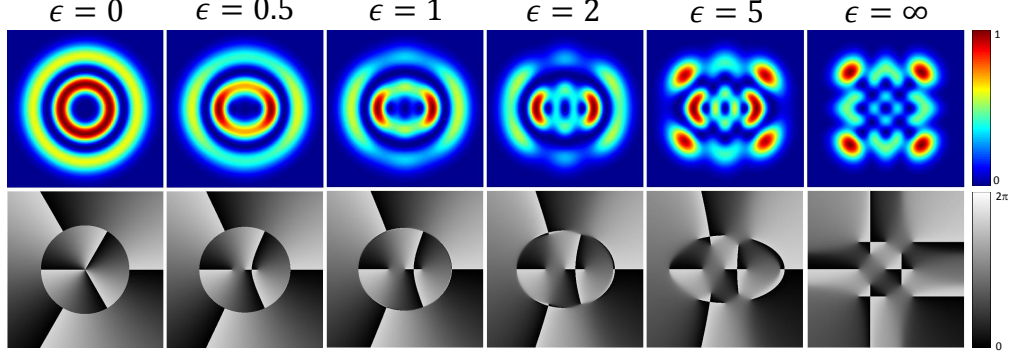


Figure 2.4: *Intensity and Phase of Ince-Gauss mode $IG_{p=5, m=3, \epsilon}$ with two rings and three vortices with varying ellipticity ϵ . For $\epsilon = 0$, the $IG_{5,3,\epsilon}$ reduces to a Laguerre-Gauss mode with $n = \frac{p-m}{2} = 1$ and $\ell = m = 3$. For increasing ellipticity, the three-fold vortex splits into three vortices of charge one. For $\epsilon \rightarrow \infty$, the modes reduces to helical Hermite-Gauss modes [25] (Figure from [14])*

2.3 On an apparent paradoxon

The content of this chapter is based on the publication “On Small Beams with Large Topological Charge” [28]³.

Laguerre-Gauss modes (as introduced in chapter 2.2.2) are frequently expressed as solutions of the paraxial wave equation. As explained in its derivation in chapter 2.1, the paraxial wave equation is a small-angle approximation of the Helmholtz equation. There, the Laguerre-Gauss modes have well-defined orbital angular momentum. The paraxial solutions predict that beams with large OAM could be used to resolve arbitrarily small distances – a dubious situation. We show how to solve that situation by calculating the properties of beams free from the paraxial approximation. We find that indeed one can resolve smaller distances with larger OAM, although with decreased visibility. If the visibility is kept constant (for instance at the Rayleigh criterion, the limit where two points are reasonably distinguishable), larger OAM does not provide an advantage. The drop in visibility is due to a field in the direction of propagation, which is neglected within the paraxial limit – which is an instructive solution to an apparent paradoxon.

Laguerre-Gauss beams with radial mode number $n = 0$ and $z = 0$ can be described in cylindrical coordinates (r, ϕ) by

$$LG_\ell(r, \phi) = N \cdot \left(\frac{r}{w_0} \right)^{|\ell|} \cdot \exp \left(-\frac{r^2}{w_0^2} \right) \cdot \exp(-i\ell\phi) \quad (2.22)$$

where w_0 is the beam waist at the focus and N is a normalisation constant. The intensity is given by

$$I_\ell(r, \phi) = |LG_\ell(r, \phi)|^2, \quad (2.23)$$

³Mario Krenn, Nora Tischler, Anton Zeilinger, On small beams with large topological charge. *New Journal of Physics* **18**, 033012 (2016).

2 Background

which results in an intensity ring for $\ell > 0$. The intensity maximum of the ring in the radial direction can be found by $\partial_r I_\ell(r, \phi) = 0$ and is

$$r_{\max} = \sqrt{\frac{\ell}{2}} w_0 \quad (2.24)$$

The radius of maximum intensity scales with the square-root of the OAM [29, 30]. Superpositions of two LG modes with opposite OAM have the same radial dependence (consequently, the intensity maximum is at the same $r_{\max}$), but they exhibit intensity oscillations in the azimuthal direction with 2ℓ intensity maxima and minima. The distance Δ between two intensity maxima is therefore

$$\Delta = \frac{\pi w_0}{\sqrt{2\ell}} \quad (2.25)$$

For a fixed beam waist, the distance between two petals becomes smaller with increasing ℓ . A potential application of these superposition beams is the use of the azimuthally varying intensity pattern to probe small structures. Then the distance Δ between two maxima will have a bearing on the achievable resolution. Based on the paraxial solution from eq.(2.22) it seems as if we should be able to resolve arbitrarily small structures since Δ can be decreased by increasing ℓ . One could even resolve in the sub-wavelength regime, which seems to contradict Abbe's diffraction limit, which gives a limit $d > \frac{\lambda}{2}$. This would obviously constitute a very curious situation and the question is how it can be resolved.

2.3.1 Non-paraxial treatment of Laguerre-Gauss modes

The calculation above is based on the paraxial approximation, which is only valid for sufficiently large beams: It is known that the paraxial wave equation is a zero-order approximation of Maxwell's equations with terms of the order $\alpha = \mathcal{O}\left(\frac{\lambda}{w_0}\right)$ ignored, where λ is the wavelength [9, 12, 31]. Here, we test if this consideration withstand a more rigorous analysis free from the paraxial approximation. We use forward-propagating LG modes which are full solutions of Maxwell's equations. In order to calculate full solutions of LG beams, we use two distinct methods proposed in the literature.

The Riemann-Silberstein Formalism

The first method is based on the elegant framework of the Riemann-Silberstein vector [32, 33]. The method treats electric and magnetic field both together in one complex vector

$$\mathbf{F} = \frac{\mathbf{E}}{\sqrt{2\epsilon_0}} + i \frac{\mathbf{B}}{\sqrt{2\mu_0}} \quad (2.26)$$

The fields can be obtained by complex scalar functions $\chi(r, t)$, which are called Hertz-potential for the beams in z-direction [33]:

$$\mathbf{F} = \begin{pmatrix} \partial_x \partial_z + \frac{i}{c} \partial_y \partial_t \\ \partial_y \partial_z - \frac{i}{c} \partial_x \partial_t \\ -(\partial_x^2 + \partial_y^2) \end{pmatrix} \chi(r, t) \quad (2.27)$$

2 Background

For LG beams, an analytical, finite energy solution has been presented in [34]:

$$\chi_{\Omega, \ell, n=0}^{\sigma}(r, \phi, z, t) = N \cdot \frac{e^{-i\sigma(\Omega(t - \frac{z}{c}) - \ell\phi)} r^{\ell}}{a(t_+)^{\ell+1}} e^{-\frac{r^2}{a(t_+)^2}} \quad (2.28)$$

with $a(t_+) = w_0^2 + i \frac{\sigma c^2(t + \frac{z}{c})}{\Omega}$, where N is a normalisation constant, σ is the circular polarisation, and Ω is the optical (mean) frequency. Applying eq.(2.28) to eq.(2.27), we find the full and finite-energy electromagnetic field. However, the resulting solution is not monochromatic: The optical frequency spreads around the central optical frequency Ω , particularly in the non-paraxial regime this effect is significant. Thus this solution is more difficult to interpret with respect to the diffraction limit as they are not monochromatic. To avoid these difficulties, we continue with the aplanatic lens model, which describes the process of focusing.

The aplanatic lens model

The aplanatic lens model can be used to calculate strongly focused fields that are obtained by the use of a microscope objective [35, 36]. Based on properties of the focusing optics and of the incident field, the model provides the focused electric field, which is a solution of the full Maxwell equations. The properties specifying the focusing optics are the focal length, the numerical aperture (NA), and the transmission coefficients for s- and p-polarization. We assume an ideal microscope objective by setting the NA to 1 and by letting the s and p transmission coefficients be unity, which is the goal of antireflection coatings. For the incident field, we use the LG modes specified by eq.(2.22) with circular polarisation

$$\hat{\mathbf{u}}(\mathbf{r}, \sigma) = \frac{1}{\sqrt{2}} (\hat{\mathbf{x}} + i\sigma \hat{\mathbf{y}}). \quad (2.29)$$

Such a circularly polarized collimated beam is a very good approximation a helicity eigenstate (with eigenvalue $\sigma = \pm 1$), and the helicity (i.e. circular polarization of each plane wave composing the total field) is preserved throughout the focusing process owing to the equal s- and p-transmission coefficients [37].

The aplanatic lens model transforms the real space field of the input beam to a plane wave decomposition of the focused field. For LG modes this is done by using a paraxial input field at the lens

$$\mathbf{E}_{in}(\mathbf{r}, t) = \mathbf{E}_{in}(r, \phi, 0, t) = \text{LG}_{\ell}(r, \phi) \cdot \hat{\mathbf{u}}(\mathbf{r}, \sigma) \quad (2.30)$$

where $\text{LG}_{\ell}(r, \phi)$ is defined in eq.(2.22) and $\hat{\mathbf{u}}(\mathbf{r}, \sigma)$ is defined in eq.(2.29). Then the coefficients of the plane wave decomposition for the focused field are obtained by making the substitutions

$$\begin{aligned} r &\rightarrow \frac{f \cdot k_r}{k} \\ \phi &\rightarrow k_{\phi} \end{aligned} \quad (2.31)$$

for the coordinates of the input field.

The new polarization vectors in the plane wave decomposition can be obtained as follows: We start with a helicity eigenstate and our aplanatic lens conserves that helicity, which means

2 Background

that each plane wave of the focused field must have circular polarization with the same handedness as the incident field. The new $\hat{\mathbf{u}}(\mathbf{k}, \sigma)$ fulfills the following requirements: It is a normalized circular-polarization vector [33] in Cartesian coordinates, which is tilted such that it is transverse to the momentum vector $\mathbf{k}$, and has its handedness specified by σ . Specifically, $\hat{\mathbf{u}}(\mathbf{k}, \sigma)$ can be derived by starting with $\hat{\mathbf{u}}(\mathbf{r}, \sigma)$, and rotating this vector by $\arcsin\left(\frac{k_r}{k}\right)$ about the y axis, and by k_ϕ about the z axis. Then an $\exp(i\sigma\ell)$ factor is applied in order to attain the correct total angular momentum $J = \ell + \sigma$, which is the total angular momentum of the input beam that is conserved by the cylindrically symmetric focusing optics. The final step consists in a Fourier transform to obtain the real space field of focussed LG modes:

$$\begin{aligned} \mathbf{E}_{\ell,\sigma}(\mathbf{r}, t) &= \int_0^{k_{\max}} g(k_r) \cdot k_r dk_r \int_0^{2\pi} dk_\phi \text{LG}(k_r, k_\phi, w_0) \cdot e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)} \cdot \hat{\mathbf{u}}(\mathbf{k}, \sigma) \\ \text{LG}(k_r, k_\phi, w_0) &= \sqrt{\frac{w_0^2}{2\pi|\ell|!}} \cdot e^{i\ell k_\phi} \cdot \left(\frac{w_0 k_r}{\sqrt{2}}\right)^{|\ell|} \cdot e^{-\frac{w_0^2 k_r}{4}} \cdot e^{-i\frac{|\ell|\pi}{2}} \\ \hat{\mathbf{u}}(\mathbf{k}, \sigma) &= \frac{e^{i\sigma k_\phi}}{\sqrt{2}k \cdot k_r} \cdot \begin{pmatrix} -k_x k_z + \sigma i k k_y \\ -k_y k_z - \sigma i k k_x \\ k_x^2 + k_y^2 \end{pmatrix} \end{aligned} \quad (2.32)$$

Here, $w_0 = \frac{f \cdot \lambda}{w_{in} \pi}$ is the beam waist after focusing the incoming beam (with waist w_{in}) with a lens of focal length f . (k_r, k_ϕ, k_z) and (k_x, k_y, k_z) are cylindrical and cartesian coordinates in momentum space, with $k = \sqrt{k_r^2 + k_z^2} = \sqrt{k_x^2 + k_y^2 + k_z^2} = \frac{2\pi}{\lambda}$ denoting the wave number and λ being the optical wavelength. The integration of k_r is cut off at $k_{\max} = k$ which implies a numerical aperture of the focusing objective of 1, and effectively avoids evanescent waves as we are only interested in propagating fields. The factor $g(k_r) = \sqrt[4]{1 - \left(\frac{k_r}{k}\right)^2}$ comes from energy flux conservation during focusing, and is responsible for a damping of high radial k-components at very strong focusing.

The helicity is given by σ , which can be +1 or -1 for left- or right-circular polarization. $\hat{\mathbf{u}}(\mathbf{k}, \sigma)$ is a normalized circular-polarization vector in Cartesian coordinates [33]. As we would like to produce superposition of LG modes, we can simply add two fundamental solutions:

$$\mathbf{E}_{\pm\ell,\sigma}(\mathbf{r}, t) = \frac{1}{\sqrt{2}} (\mathbf{E}_{+\ell,\sigma}(\mathbf{r}, t) + \mathbf{E}_{-\ell,\sigma}(\mathbf{r}, t)) \quad (2.33)$$

2.3.2 Applications to small Laguerre-Gauss modes

Now we study the intensity distribution of very small beams with non-zero OAM superposition. The intensity can simply be calculated as

$$I_\ell(r, \phi) = \sum_{i=\{x,y,z\}} \text{Re}(E_i)^2 + c^2 \text{Re}(B_i)^2, \quad (2.34)$$

where $\text{Re}()$ denotes the real part. The intensity $I_\ell(r, \phi)$ is related to the full energy of the electromagnetic field. It is also meaningful in quantum physics due to its interpretation as the probability density for the case of single photons [33, 38]. We fix our wavelength to $\lambda=800\text{nm}$. In figure 2.5 we plot the intensity of a beam with $\ell=15$, which leads to 30 petals in the ring. In Fig. 2.5A-B, the beam waist is $w_0=10 \mu\text{m}$, whereas in Fig. 2.5C-D $w_0=1 \mu\text{m}$ is used. In

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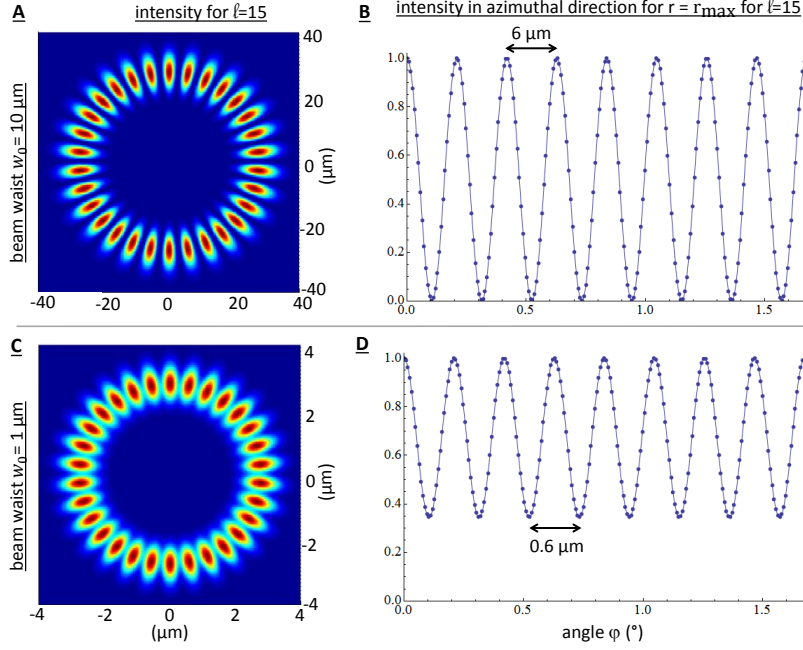


Figure 2.5: LG beams with $\lambda=800\text{nm}$ and $\ell=15$ with different beam waist w_0 . A: intensity profile of the LG mode with $w_0=10\mu\text{m}$. B: Intensity in azimuthal direction for $r=r_{\text{max}}$. The visibility of the fringes is close to unity, which is very close to the paraxial case. C & D: The same properties for a LG beam with $w_0=1\mu\text{m}$. The minima are washed out significantly, the visibility drops to roughly 49%, which is a pure non-paraxial effect.

the smaller beam, the intensity minima are significantly filled in due to a large field component in z-direction (in the example, the maximum of the field in z-direction is roughly 32% of the full intensity's maximum). The z-component is shifted azimuthally by exactly half a period compared to the x- and y-components. The visibility, defined as

$$\text{vis} = \frac{I_{\text{max}} - I_{\text{min}}}{I_{\text{max}} + I_{\text{min}}}, \quad (2.35)$$

at the radial position of the intensity maximum, drops from almost unity to 48.6%. In figure 2.6, we show the OAM dependence of the beam waist w_0 , the maximum radius r_{max} and the distance between different maxima Δ . If no other restrictions are applied and w_0 remains constant, indeed r_{max} scales like the square root of the OAM, therefore Δ decreases - exactly as predicted by the paraxial solution. However, if one takes into account the decrease of the visibility, which is a significant measure for resolution, the situation changes. When the visibility is fixed (by adjusting w_0), the maximum radius r_{max} increases linearly with the OAM, which leads to a constant behaviour of the Δ . The behavior is analyzed for different visibilities of superposition fringes: 95%, 80%, 50% and 15.1%. The last one resembles the Rayleigh criterion, the limit at which two points can be reasonably distinguished. These results show that OAM superpositions cannot be used to decrease the distance between two maxima while keeping the minima small.

These conclusions are based on the natural choice of using the radius at which the total

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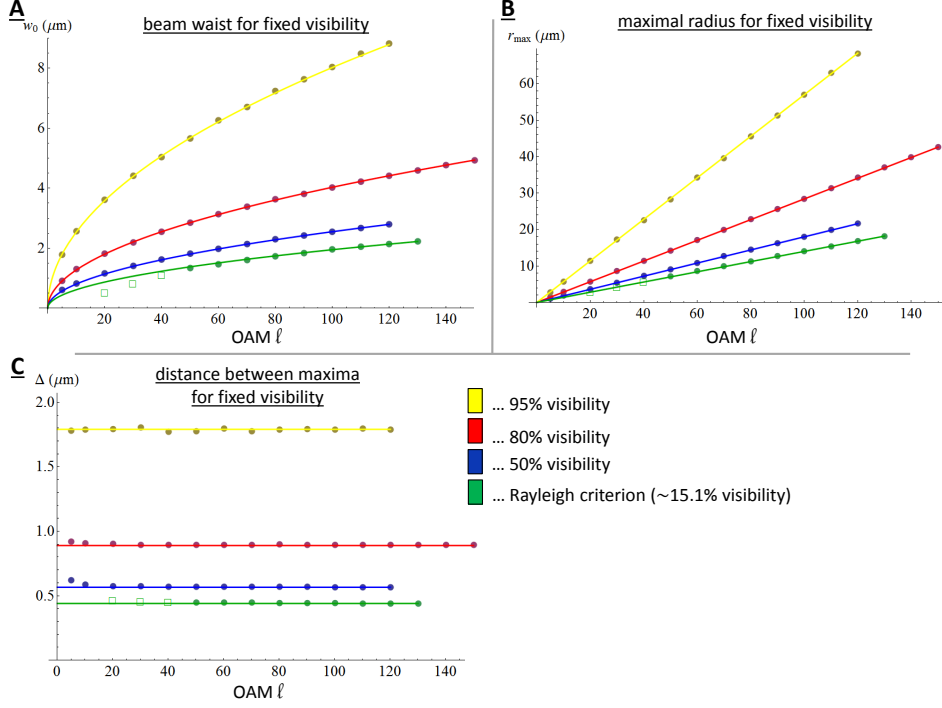


Figure 2.6: A: If we fix the visibility of the intensity in the azimuthal direction, we find that the beam waist w_0 increases as a square root of the OAM. B: This leads to a linear scaling of the maximum radius of the intensity pattern. C: As a consequence, the distance Δ stays constant. For all plots the points represent calculated values and the line stands for square-root or linear interpolation. The cases where significant portions of the beam are cut off due to the cutoff k_{max} from eq.(2.33) are indicated by squares.

energy of the field is maximal. One interesting matter is the amplitude of the fringes. In all cases presented above we have analyzed the visibility in the radial position of maximal intensity. We also analyzed azimuthal visibilities for $r \neq r_{\text{max}}$, and see that the visibility decreases for smaller r (in regions where there is still a considerable amount of intensity). This is interesting as situations can exist where high frequency oscillations with perfect visibilities can be achieved, in places where the intensity is exponentially small [39].

In figure 2.7, we calculate the visibilities for different regions of the beam, for different OAM ℓ and different beam waists w_0 . The visibility is calculated for all radii where the intensity is at least 0.1% of the maximum intensity - excluding regions of negligible intensity, which are expected to be unusable in imaging applications. We find that for smaller radii, the visibility only decreases. For larger radii, it increases. In order to calculate the visibility in other regimes where the intensity is even lower, robust numerical methods need to be used in order to deal with exponentially small intensities.

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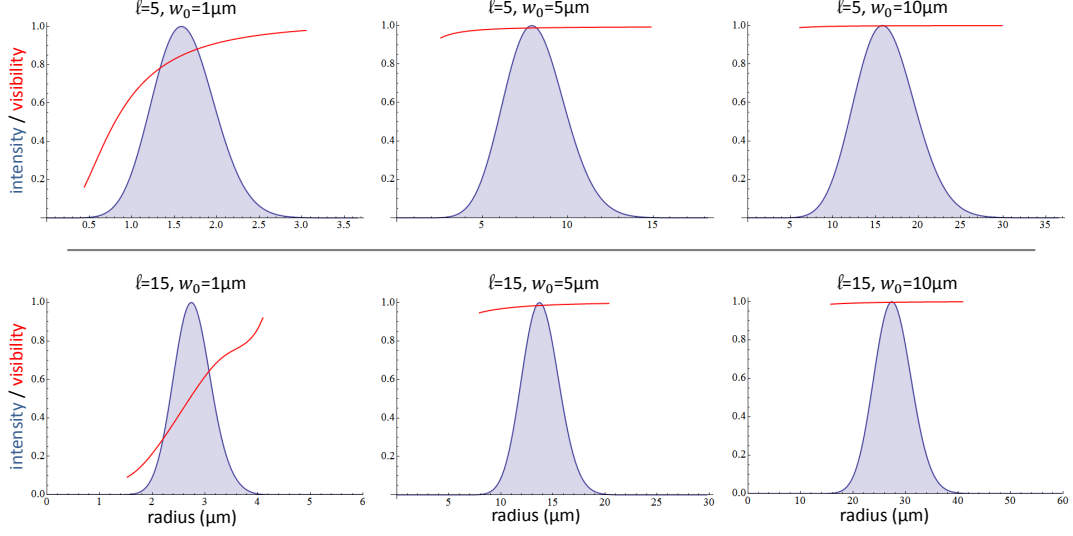


Figure 2.7: The visibility of beams with different waists and different OAM is calculated as a function of the radial coordinate (not only at the radius of maximum intensity - as used in the main text). The blue shape represents the intensity of the LG mode in radial direction. The red line shows the visibility.

2.3.3 Remarks

The careful non-simplified treatment shows that the paraxial approximation eq.(2.22) leads to incorrect predictions in the regime of small beams with large ℓ . This is for two distinct reasons. First of all, the field component in the z -direction, which is neglected in the paraxial approximation, causes reduced visibilities, with possible implications for imaging applications or optical lattices [40]. Secondly, limiting the field to propagating modes imposes a cutoff in the radial momentum components, which becomes significant for even smaller beam waists or larger ℓ (see eq.(2.33)).

A fascinating question is the behaviour of matter waves with large orbital angular momentum [41–43]. As they are described by the Schrödinger equation, which has the same form as the paraxial wave equation, the visibility issue in OAM superpositions do not apply - indicating an interesting difference between propagation of photons [44, 45] and matter waves [46, 47]. However, the physical constraint on the maximal transverse momentum for propagating modes is valid for matter waves and poses a limitation on their use for resolution applications, similarly as in the case of photons. The paraxial wave equation, for which eq.(2.22) is a solution, is an approximation of an optical Dirac equation [48], in a formally very similar way as the Schrödinger equation is an approximation of the Dirac equation. It would be interesting to investigate whether similar non-paraxial effects presented here exist in some form for relativistic matter waves as well.

2.4 Quantum States and Mutually Unbiased Bases

Single photons can be in a coherent superposition of two or more orthogonal states. Such states are called qubit or qudit, respectively. Qubits can be fully described using two real numbers, where one describes the amplitude and the other stands for the relative phase. Let's consider $|0\rangle$ and $|1\rangle$ as two orthogonal quantum states (which are defined in an *Hilbert space*), then a qubit is written as

$$|\psi\rangle = \cos\left(\frac{\theta}{2}\right) |0\rangle + e^{i\phi} \sin\left(\frac{\theta}{2}\right) |1\rangle \quad (2.36)$$

where the $0 \leq \theta \leq \pi$ and $0 \leq \phi < 2\pi$. A qubit can conveniently be depicted at a *Bloch sphere*, a unity 2-dimensional sphere as shown in figure (2.8).

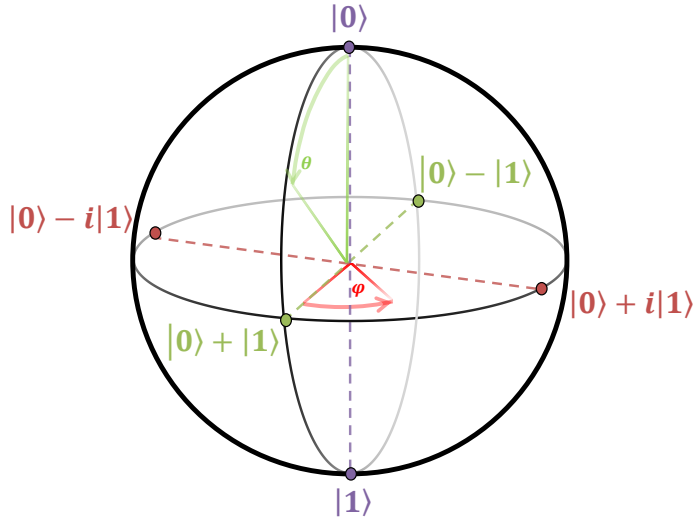


Figure 2.8: The Bloch sphere is a graphical representation of a qubit. ϕ is the polar angle which defines the relative phase between $|0\rangle$ and $|1\rangle$, and θ is the azimuthal angle which defines the relative weighting or amplitude of the two components.

There are three complete and orthogonal bases in a qubit system, which are mutually unbiased – the computation basis with the basis vectors, as well as two superposition bases (different sets of MUBs can be obtained by rotation of the Bloch sphere in Fig. 2.8)

$$\mathcal{B}_1 = \{|0\rangle, |1\rangle\} \quad (2.37)$$

$$\mathcal{B}_2 = \left\{ \frac{|0\rangle + |1\rangle}{\sqrt{2}}, \frac{|0\rangle - |1\rangle}{\sqrt{2}} \right\} \quad (2.38)$$

$$\mathcal{B}_3 = \left\{ \frac{|0\rangle + i|1\rangle}{\sqrt{2}}, \frac{|0\rangle - i|1\rangle}{\sqrt{2}} \right\} \quad (2.39)$$

with the property

$$\forall(k, l, i) : |\langle v_{\mathcal{B}_i}^k | v_{\mathcal{B}_i}^l \rangle|^2 = \delta_{k,l}. \quad (2.40)$$

For *mutually unbiased bases* (MUBs), the overlap between every basis vector of basis $\mathcal{B}_i$ and $\mathcal{B}_j$ is one-half:

$$\forall(k, l) \& (i \neq j) : |\langle v_{\mathcal{B}_i}^k | v_{\mathcal{B}_j}^l \rangle|^2 = \frac{1}{2}, \quad (2.41)$$

2 Background

where $v_{\mathcal{B}_i}^k$ is the k -th basis vector of the basis $\mathcal{B}_i$. This extraordinary property means that one can encode one bit of information in either of the three basis, and can retrieve it in the same basis. However, if one chooses to analyse the encoded qubit in a different basis (which is unbiased to the first one), the outcome is completely random and one has no information about the originally encoded state.

The Hilbert space describing the quantum state can have more than two basis vectors. In such cases, one talks about a *high-dimensional Hilbert space*. The computation basis can always be written as

$$\mathcal{B}_1 = \{|0\rangle, |1\rangle, \dots, |d-1\rangle\}, \quad (2.42)$$

and mutually unbiased bases can be generalized in higher dimensions in the following form:

$$\forall(k, l) \& (i \neq j) : |\langle v_{\mathcal{B}_i}^k | v_{\mathcal{B}_j}^l \rangle|^2 = \frac{1}{d}, \quad (2.43)$$

where again $v_{\mathcal{B}_i}^k$ is the k -th d -dimensional basis vector of the basis $\mathcal{B}_i$. Surprisingly, it is not known in general, how many mutually unbiased bases exist for dimensions d . For dimensions $d = p^n$ (with p being a prime number, and $n \in \mathbb{N}$), one can always find $(d+1)$ MUBs (see for instance [49] for the construction of MUBs up to $d=9$). For dimension which are not powers of primes, e.g. $d = 6 \cdot 2 \cdot 3$, it is not known how many such bases exist. For $d=6$, at the moment three MUBs are known. This leads to the curious situation that in five dimensions, there are more independent ways to encode information than in the (larger) six-dimensional space.

2.5 Two-Party Quantum States and Quantum Entanglement

The content of this section is partially based on the publications [50]⁴ and [51]⁵

The general two-qubit state $|\psi\rangle$ (where particle A and B live in a Hilbert space $\mathcal{H}_A$ and $\mathcal{H}_B$ respectively) is described in the combined Hilbert space of $\mathcal{H} = \mathcal{H}_A \otimes \mathcal{H}_B$.

$$|\psi\rangle = a_{0,0}|0_A, 0_B\rangle + a_{0,1}|0_A, 1_B\rangle + a_{1,0}|1_A, 0_B\rangle + a_{1,1}|1_A, 1_B\rangle, \quad (2.44)$$

where $a_{i,j} \in \mathbb{C}$ are the complex amplitudes of the state. Quantum states which can be described as product states are called *separable* and can be written as

$$|\psi\rangle_{sep} = |\psi\rangle_A \otimes |\psi\rangle_B. \quad (2.45)$$

Examples of separable states are

$$|\psi\rangle_{sep} = |0_A, 0_B\rangle \quad (2.46)$$

$$|\psi\rangle_{sep} = \frac{1}{2} (|0_A, 0_B\rangle + |0_A, 1_B\rangle + |1_A, 0_B\rangle + |1_A, 1_B\rangle) \quad (2.47)$$

$$= (|0\rangle + |1\rangle)_A \otimes (|0\rangle + |1\rangle)_B. \quad (2.48)$$

⁴Mario Krenn, Mehul Malik, Thomas Scheidl, Rupert Ursin, Anton Zeilinger, Quantum communication with photons. *Optics in our Times* (Springer International Publishing, 2016) p. 455-482.

⁵Mario Krenn, Mehul Malik, Manuel Erhard, Anton Zeilinger, Orbital angular momentum of photons and the entanglement of Laguerre–Gaussian modes. *Phil. Trans. R. Soc. A* **375**, 20150442 (2017).

2 Background

One of the most curious properties of quantum systems is that it is not always possible to describe the properties of $|\psi\rangle$ just by the local properties of particle A and B independently. Those quantum states which are not separable as defined in equation (2.45) are called *entangled states*. One example is the following

$$|\psi\rangle_{ent} = \frac{1}{\sqrt{2}}(|0_A, 0_B\rangle + |1_A, 1_B\rangle) \quad (2.49)$$

which is a coherent superposition between both particles in the state $|0\rangle$ and both particles in state $|1\rangle$. Before we measure the state, the two photons do not have a well-defined value of 0 or 1. Only if we measure the state in the $\{|0\rangle, |1\rangle\}$ basis, one of the two possibilities is realized at random. The *randomness* does not only come into play because of our limited knowledge but is a phenomenon of the quantum mechanical description, as we can see in the following simple example: Let us consider a polarization-entangled photon pair, where $|0\rangle$ is horizontal (H) polarisation, and $|1\rangle$ is vertical (V) polarisation. If the two particles would have a well-defined value of being either in H or V , we could describe them with a classical mixture:

$$\rho_{classical} = \frac{1}{2}(|H, H\rangle\langle H, H| + |V, V\rangle\langle V, V|) \quad (2.50)$$

which would lead to 50 % of counts where both photons are H , and 50 % where they are V . However, if one measures the photons in a superposition basis (2.38), which corresponds to diagonal (D) and antidiagonal (A) polarisation, one finds

$$\rho_{classical} = \frac{1}{4}(|D, D\rangle\langle D, D| + |D, A\rangle\langle D, A| + |A, D\rangle\langle A, D| + |A, A\rangle\langle A, A|) \quad (2.51)$$

which means that all four possible combinations would occur. In contrast, a measurement of the entangled state (2.49) will lead to

$$|\psi\rangle_{ent} = \frac{1}{\sqrt{2}}(|D, D\rangle + |A, A\rangle). \quad (2.52)$$

Thus we see, the assumption that the two particles have well-defined values prior to the measurements leads to a contradiction. The formalism does not contain any space- or time-information – that means that particle A and B can be separated by a large distance, and still a measurement of particle A would immediately set the value of particle B . One can freely choose the measurement basis of the state, thus the particle A will be realized as an eigenstate in that basis. Furthermore, also its partner particle B will be realized immediately as an eigenstate in that basis. Couldn't that be used to communicate faster than speed of light?

Such a protocol has been proposed in 1982 [52], and exploits the fact that the basis of the entangled photon can be changed faster than speed of light. It requires the usage of a *quantum cloning machine* because the basis can not be decided with a single photon only, see figure (2.9). Unfortunately, there is one problem with that protocol: It cannot work. Already in the same year in 1982, Wootters and Zurek found that quantum mechanics forbids to perfectly clone a quantum state [53] (related properties have already been discussed by Stephen Wiesner in 1970 [54]). This profound result originates from a simple property of quantum mechanics, namely the linear superposition principle. We can inspect what a potential cloning-operation $\hat{C}$ would do. We use an input quantum state, and an undefined second photon $|X\rangle$. After the cloning operation, the second photon should have the polarisation property of the first photon. This is how our cloning machine would act on states in the H/V-basis:

2 Background

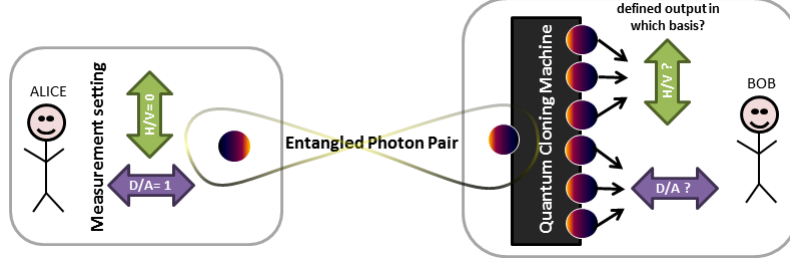


Figure 2.9: Visualisation of a faster-than-light quantum communication protocol, if (!) quantum states could be cloned: Alice and Bob share an entangled photon pair. By choosing the measurement basis between horizontal/vertical or diagonal/antidiagonal polarisation, Alice projects the whole state into an eigenstate of that basis. This means that Bob's state is also an eigenstate in that basis. To find the basis chosen by Alice, Bob would need to measure more than one photon. He can do that by observing many photons in both the H/V basis and the D/A basis. The basis in which the photon is an eigenstate will lead to 100% counts in one of the two possibilities and 0% in the other, while the wrong basis will give 50% of the counts in either of the possibilities. If he could perfectly clone his photon, he could find the basis (by repeating the measurement many times and compare the two statistics), and receive the information faster than light. Unfortunately, this is prohibited by the no-cloning theorem, a fundamental rule in quantum mechanics. (Figure from [50])

$$\hat{C}(|H\rangle |X\rangle) = |H\rangle |H\rangle \quad (2.53)$$

$$\hat{C}(|V\rangle |X\rangle) = |V\rangle |V\rangle \quad (2.54)$$

The cloning-machine should work in every basis, thus we inspect what happens when we try to clone a diagonally polarised photon $|D\rangle$. Note that a diagonally polarised photon can be expressed in the H/V basis as a coherent superposition of a horizontal and a vertical part $|D\rangle = \frac{1}{\sqrt{2}}(|H\rangle + |V\rangle)$. The quantum cloning machine acts as

$$\begin{aligned} \hat{C}(|D\rangle |X\rangle) &= \\ &= \hat{C}\left(\frac{1}{\sqrt{2}}(|H\rangle + |V\rangle) |X\rangle\right) \\ &= \frac{1}{\sqrt{2}}(\hat{C} |H\rangle |X\rangle + \hat{C} |V\rangle |X\rangle) \\ &= \frac{1}{\sqrt{2}}(|H\rangle |H\rangle + |V\rangle |V\rangle) \end{aligned} \quad (2.55)$$

The last line in equation (2.55) was obtained by using equations (2.53) and (2.54) for the cloning operator $\hat{C}$. The result is an entangled state that cannot be factored into $|D\rangle |D\rangle$. If one were to measure either of the entangled photons individually, the result would be random, and certainly not $|D\rangle$. From this simple example it is clear that quantum cloning is not possible. This property prohibits faster-than-light communication, but it opens the door to many different quantum secret sharing protocols, such as quantum cryptography.

2 Background

2.5.1 Bell basis

An exceptionally useful basis for quantum information tasks is the basis of maximally entangled two-particle states, also known as the Bell basis. It consists of four orthogonal states

$$|\phi^+\rangle = \frac{1}{\sqrt{2}}(|0,0\rangle + |1,1\rangle) \quad (2.56)$$

$$|\phi^-\rangle = \frac{1}{\sqrt{2}}(|0,0\rangle - |1,1\rangle) \quad (2.57)$$

$$|\psi^+\rangle = \frac{1}{\sqrt{2}}(|0,1\rangle + |1,0\rangle) \quad (2.58)$$

$$|\psi^-\rangle = \frac{1}{\sqrt{2}}(|0,1\rangle - |1,0\rangle). \quad (2.59)$$

Every (pure) two-particle two-dimensional quantum state can be represented in terms of superpositions of Bell states – independent of whether it is entangled or not. For example, the two separable states in (2.46) and (2.48) can be written as

$$|\psi\rangle_{sep} = |0_A, 0_B\rangle \quad (2.60)$$

$$= \frac{1}{\sqrt{2}}(|\phi^+\rangle + |\phi^-\rangle) \quad (2.61)$$

$$|\psi\rangle_{sep} = \frac{1}{2}(|0_A, 0_B\rangle + |0_A, 1_B\rangle + |1_A, 0_B\rangle + |1_A, 1_B\rangle) \quad (2.62)$$

$$= \frac{1}{\sqrt{2}}(|\phi^+\rangle + |\psi^+\rangle). \quad (2.63)$$

That property allows for applications such as quantum teleportation [55] or entanglement swapping [56].

2.5.2 High-dimensional entanglement

High-dimensional two-particle systems can also be correlated. In this case, there are three different cases: correlations in a high-dimensional space, entanglement in a high-dimensional space and genuine high-dimensional entanglement.

A classically correlated state in three dimensions is for example

$$\rho_{1d} = \frac{1}{3}(|0,0\rangle\langle 0,0| + |1,1\rangle\langle 1,1| + |2,2\rangle\langle 2,2|). \quad (2.64)$$

Such states can be produced by a classical source that produces two photons carrying $\ell = 0$ sometimes, and $\ell = 1$ or $\ell = 2$ at other times, resulting in a completely incoherent mixture. For example, two-dimensional entangled states can be written in the following way:

$$\begin{aligned} |\psi_{2d}^{0,1}\rangle &= \frac{|0,0\rangle + |1,1\rangle}{\sqrt{2}} \\ |\psi_{2d}^{0,2}\rangle &= \frac{|0,0\rangle + |2,2\rangle}{\sqrt{2}} \\ |\psi_{2d}^{1,2}\rangle &= \frac{|1,1\rangle + |2,2\rangle}{\sqrt{2}}, \end{aligned} \quad (2.65)$$

An incoherent mixture between these three states,

$$\rho_{2d} = \frac{1}{3}(|\psi_{2d}^{0,1}\rangle\langle\psi_{2d}^{0,1}| + |\psi_{2d}^{0,2}\rangle\langle\psi_{2d}^{0,2}| + |\psi_{2d}^{1,2}\rangle\langle\psi_{2d}^{1,2}|), \quad (2.66)$$

2 Background

has correlations in three dimensions, however it is still only two-dimensional entangled, i.e. its Schmidt number is two. The important challenge is to distinguish it from genuine three-dimensional entangled states such as

$$|\psi_{3d}\rangle = \frac{|0,0\rangle + |1,1\rangle + |2,2\rangle}{\sqrt{3}} \tag{2.67}$$

We will come back to this question in chapter (3).

3 Quantum Entanglement in 100 dimensions

The content of this chapter is based on the publication "*Generation and confirmation of a (100×100)-dimensional entangled quantum system*" [57]^{1 2}.

3.1 Motivation

In this chapter, a high-dimensional two-photon quantum system is investigated experimentally. The photons are high-dimensionally entangled in transverse spatial modes. We investigate the non-classical correlation in the discrete Laguerre-Gauss basis, which has been introduced in chapter (2.2.2). The dimensionality of the entanglement is measured using a new entanglement dimensionality witness (Schmidt number witness), which only requires experimentally feasible two-dimensional subspace measurement. We find that the system is entangled in at least 100 dimensions, which is comparable to size of the Hilbert space of a 13-qubit entangled state. Entangled quantum systems have properties that have overthrown the classical worldview. As explained in chapter (2.4), the properties of such systems can not be described prior to and independent of the measurement. This counterintuitive property leads to a manifold of quantum information applications, such as entanglement-based quantum cryptography or quantum computation. To obtain a better understanding of entanglement itself, as well as its application in various quantum information tasks, increasing the complexity of entangled systems is important. Essentially, this can be done in two ways: The first method is to increase the number of particles involved in the entanglement [58–61]. The alternative method is to increase the entanglement dimensionality of a system – the method we are investigating here.

High-dimensional entanglement provides a higher information density than conventional two-dimensional (qubit) entangled states, which has important advantages in quantum communication: Firstly, it can be used to increase the channel capacity via superdense coding [62]. Secondly, high-dimensional entanglement enables the implementation of quantum communication tasks in regimes where mere qubit entanglement does not suffice. This involves situations with a high level of noise from the environment [63, 64] or quantum cryptographic systems where an eavesdropper has manipulated the random number generator involved [65]. Moreover, the entangled dimensions of the whole Hilbert space also play a interesting role in

¹Mario Krenn, Marcus Huber, Robert Fickler, Radek Lapkiewicz, Sven Ramelow, Anton Zeilinger, Generation and confirmation of a (100×100)-dimensional entangled quantum system. *PNAS* **111**, 6243–6247 (2014).

²The background shows the recorded intensity of a Laguerre-Gaussian mode superposition: $\psi = |LG_{n=0,\ell=3}\rangle + |LG_{n=2,\ell=0}\rangle$. It is one of the superpositions which were measured in the experiment explained in this chapter.

3 Quantum Entanglement in 100 dimensions

quantum computation: High-dimensional systems can be used to simplify the implementation of quantum logic [66]. A detailed example, namely a fault-tolerant quantum algorithm running on a trinary quantum computer, has been investigated and it was found that it outperforms binary architectures in terms of circuit complexity [67].

So far, high-dimensional entanglement has been implemented only in photonic systems. There, different multi-level degrees of freedom such as spatial modes [68], time-energy [69], path [70, 71] as well as continuous variables [72–74] have been employed. Especially entanglement of transverse spatial modes of photons has attracted much attention in recent years [75–84], because it is readily available from optical nonlinear crystals and because the dimension of the entanglement can be in principle very high [85].

One main challenge that remains is the detection and verification of high-dimensional entanglement. For reconstructing the full quantum state via state tomography, the number of required measurements is impractical even for relatively low dimensions because it scales quadratically with the quantum system dimension [80, 83]. Even if one had reconstructed the full quantum state, the quantification of the entangled dimensions is a daunting task, analytically and even numerically [86]. If the full density matrix of the state is not known, it is only possible to give lower bounds of the entangled dimensions. Such methods are usually referred to as a *Schmidt number witness* [87–89].

3.2 High-dimensional entanglement witness

In our experiment we are in a regime where it is unfeasible to reconstruct the full density matrix because of the required number of measurements due to the large number of parameters. But we can identify lower bounds of the number of entangled dimensions. Furthermore, all previously published methods for extracting the dimension of entanglement turned out to be impractical for our system. They usually require access to observables on the full Hilbert space, which were not available for our experiment, thus we were required to develop a novel approach.

We found a mathematically well-defined and intuitively reasonable method that answers the following question: For a given high-dimensional two-photon state, if correlations between D dimensions of each photon are measured, what is the minimum necessary entanglement dimensionality $\mathbf{d}$ required to explain the correlations? More formally, a d -dimensional entangled state has correlations that can never be explained by a convex combination of pure states with a Schmidt rank lower than d .

There is a crucial difference between *entanglement in a high-dimensional space* and *genuine high-dimensional entanglement*. A state that is entangled in a 3-dimensional space could look like

$$\rho = \frac{1}{3} \left(|\psi_{1,2}\rangle\langle\psi_{1,2}| + |\psi_{2,3}\rangle\langle\psi_{2,3}| + |\psi_{3,1}\rangle\langle\psi_{3,1}| \right) \quad (3.1)$$

with 2-dimensionally entangled pure states

$$|\psi_{m,n}\rangle = \frac{1}{\sqrt{2}} \left(|m, m\rangle + |n, n\rangle \right) \quad (3.2)$$

The state ρ in (3.1) is a incoherent mixture of three two-dimensionally entangled states,

thus ρ is two-dimensionally entangled. Even though it lives in a three-dimensional space, three-dimensional correlations are not present.

Our approach works such that we define a measurable witness-like quantity W and search for the d -dimensional entangled state maximizing it. When we perform the measurement and exceed the maximal value, we know that the measured quantum state was at least $(d+1)$ -dimensional entangled. This approach is a generalization of conventional entanglement witnesses, which define a boundary between separable and entangled states. We not only want a boundary between separable and entangled states, but also between different dimensions of entanglement.

3.2.1 Two-dimensional subspace entanglement witness

The main idea is to look at 2-dimensional subspaces, and therewith measure the correlations of the two photons. In analogy to two-dimensional systems (such as photon's polarization), we can measure the visibilities in three mutually unbiased bases (MUBs, for polarization this would be horizontal/vertical, diagonal/antidiagonal and left/right) - see figure 3.1.

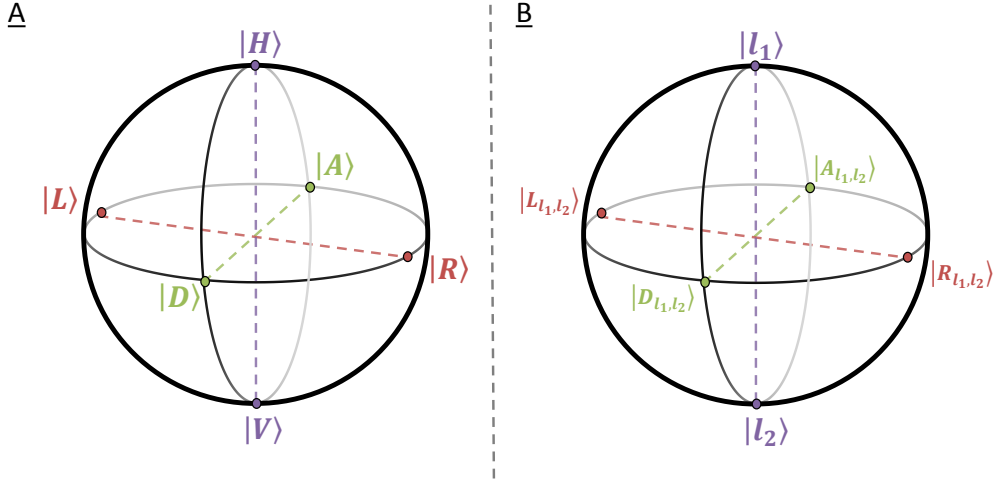


Figure 3.1: Bloch sphere of two-dimensional degrees of freedom. **A:** Polarisation, the three colors represent the three mutual unbiased bases in two dimensions. **B:** The analog OAM Bloch sphere, for the two-dimensional subspace built up by ℓ_1 and ℓ_2 . Here, $|D/A_{\ell_1, \ell_2}\rangle = \frac{1}{\sqrt{2}} (|\ell_1\rangle \pm |\ell_2\rangle)$ and $|L/R_{\ell_1, \ell_2}\rangle = \frac{1}{\sqrt{2}} (|\ell_1\rangle \pm i|\ell_2\rangle)$.

Mathematically, the visibility is defined as $V_i = |\langle \sigma_i \otimes \sigma_i \rangle|$ with $i = x, y, z$, where σ_i denote the single-qubit Pauli matrices (and x,y,z are the H/V, D/A, L/R basis for polarisation). The concept of the measurement is illustrated in figure 3.2. With these measurements, it is possible to detect entanglement between two-dimensional subsystems [89]. What we used is a way how such measurements in all two-dimensional subsystems imply a lower bound of the entangled dimensions in the whole quantum system.

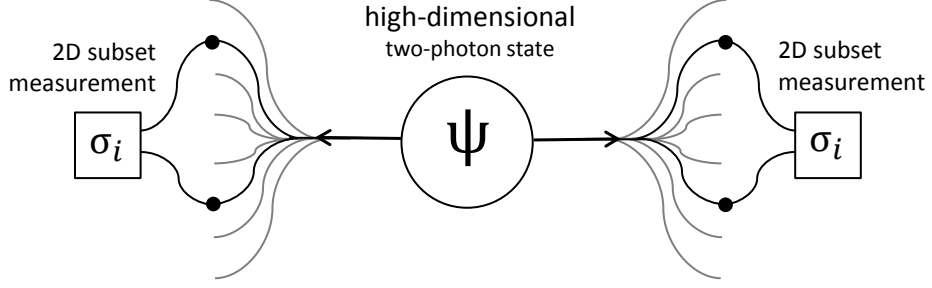


Figure 3.2: Visualization of the measurement concept. The two photons are sent into two different directions. Each of the photons is in a mixture of many modes. We perform the same two-dimensional subspace measurement on both photons. When we consider all two-dimensional subspaces, we can determine the dimensionality of entanglement.

Our entanglement witness W is the sum of the visibilities in all two-dimensional subspaces

$$W = \sum_{a=0}^{D-2} \sum_{b=a+1}^{D-1} \frac{V_x^{a,b} + V_y^{a,b} + V_z^{a,b}}{N_{a,b}} \quad (3.3)$$

where a and b stand for specific states of the photons, D is defined as above (it stands for the number of modes considered), $V_i^{a,b}$ stands for the visibility in basis i , and $N_{a,b}$ stands for the normalization. $N_{a,b}$ is the source of the nonlinearity of W which leads to convenient experimental properties, however makes it very difficult in general to handle mathematically. That nonlinearity is responsible for the fact that the measurement results are automatically normalized (i.e., all visibilities can go up to 1), because by measuring in two-dimensional subspaces, we ignore all other modes. Therefore, we do not need to renormalize our measurement results in any way afterward. Nonlinear entanglement witnesses have already been used in earlier experiments and demonstrated specific advantages over linear witnesses [27, 90].

The next step is to find the d -dimensional entangled state which is maximizing the quantity W in equation (3.3). The maximization of the witness has been performed by Marcus Huber, co-author of [57]. It is based on a combination of the method of Lagrange multipliers and algebraic considerations, and can be found in the supplementary material of [57]. Due to the nonlinearity of the witness, it was not yet possible to find the maximum for general states. However, the maximization of W was performed for a very large and particularly important class which is sufficient for our experiment. In other words, we used a physical assumption about our state in the derivation, which we will explain in more detail later in this section. This enables us to find the maximizing d -dimensional quantum state for the quantity W , and implies an upper bound on the quantity in equation (3.3) for d -dimensional entangled states, which can be written as

$$W \leq 3 \frac{D(D-1)}{2} - D(D-d) \quad (3.4)$$

3 Quantum Entanglement in 100 dimensions

If the measurements exceed the bound, the quantum state was at least $(d + 1)$ -dimensionally entangled. Otherwise, if the inequality is fulfilled (W is smaller than the right side of equation 3.4), we cannot make a statement about the dimensionality of entanglement.

The bounds can be understood intuitively: A maximally entangled state in D dimensions will have a visibility of one in all three MUBs in every two-dimensional subspace. This is represented by the first term on the right side. If the entanglement dimensionality of the state is smaller than that of the measured Hilbert space, the maximally reachable value decreases by D for each non-entangled dimension ($D - d$), which is expressed by the second term.

As an example, we consider a three-dimensional space spanned by $|0\rangle$, $|1\rangle$ and $|2\rangle$. As our space is three-dimensional, $D = 3$. In a three-dimensional space, we have three two-dimensional subspaces, see figure 3.3. In each of these subspaces, we can measure visibilities in three MUBs.

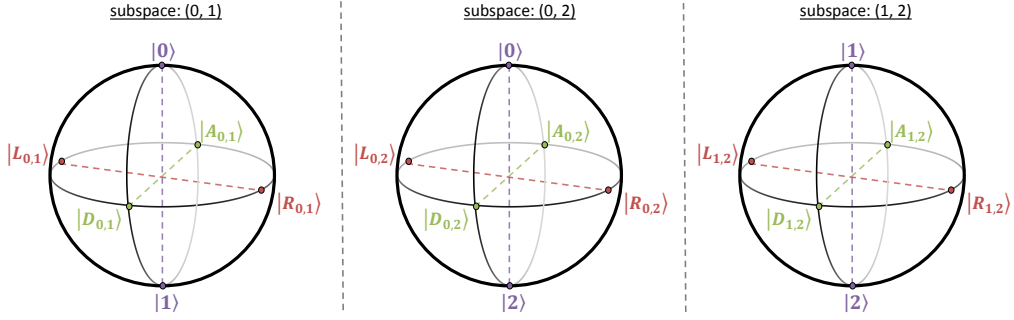


Figure 3.3: These are Bloch spheres of the three two-dimensional subspaces of $(0,1,2)$. In each of these Bloch spheres, we find three MUBs. Thus, we can measure nine visibilities.

For values of $d=(1,2,3)$, we can find a bound of W corresponding to an separable state (for $d=1$: *one-dimensional entangled*=separable), two-dimensional entanglement and three-dimensional entanglement:

$$W : \begin{cases} W \leq 3 : \text{separable} \\ 3 < W \leq 6 : \text{at least two-dimensional entangled} \\ 6 < W \leq 9 : \text{at least three-dimensional entangled} \end{cases} \quad (3.5)$$

Interestingly, the state in equation (3.1), which is defined in a three-dimensional state, could theoretically lead to $W = 6$, which is exactly the bound for two-dimensionally entangled states.

3.3 Experimental implementation

In our experiment, we apply the entanglement witness in eq.(3.5) to a two-photon quantum system. The photon pair is created by pumping a nonlinear crystal with a laser, where spontaneous parametric down-conversion (SPDC) occurs. For the high-dimensional degree of freedom we use spatial modes of light. Specifically, we use the Laguerre-Gauss (LG) basis to analyze entanglement. As described in chapter 2.2.2, LG modes form a basis of solutions of the paraxial wave equation in the cylindrical coordinate system. They are described by two quantum

3 Quantum Entanglement in 100 dimensions

numbers. One quantum number ℓ corresponds to the orbital angular momentum (OAM, or equivalently, the topological charge) of the photon [1, 3], the other is associated with the number of phase-jumps in radial direction. Photon pairs from SPDC are momentum entangled. Since OAM modes are superpositions of momentum eigenstates, it gives an intuitive reason why OAM modes are entangled as well. Of course, a rigorous treatment is more involved, see for example [91].

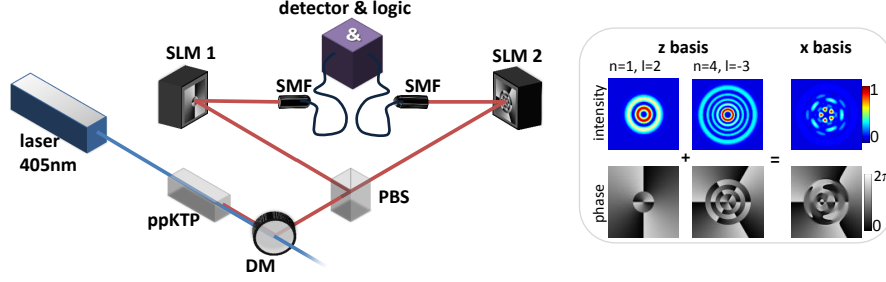
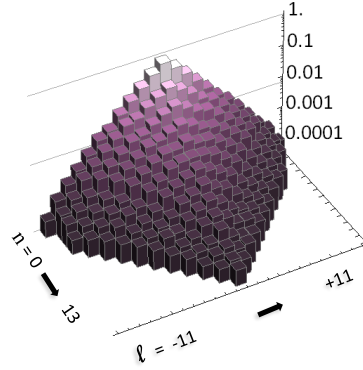


Figure 3.4: Schematic of the experimental setup. We pump a type-II nonlinear periodically poled potassium titanyl phosphate (ppKTP) crystal with a 405-nm, 40-mW single-mode laser. SPDC creates collinear photon pairs with 810-nm wavelength and orthogonal polarizations. We remove the pump beam at a dichroic mirror (DM) and separate the two photons at a polarizing beam splitter (PBS). In both arms of the setup we use SLMs to perform a mode transformation of the photons. The transformation done by a computer-generated hologram at the SLM converts a specific mode into the fundamental Gauss mode. Only the Gauss mode couples into an single mode fiber (SMF), thus the SLM + SMF combination acts as a mode filter [3]. In the end, we detect the photons with avalanche photodiode based single-photon detectors and analyze the time correlation using a coincidence logic. Left: An example of a two-dimensional subspace is shown. The intensities and phases for two different modes in the z basis are demonstrated, and their superposition leads to a mode in the x basis. The y basis can be constructed similarly.

a Normalized Count rates for involved modes



b Correlations in computational basis

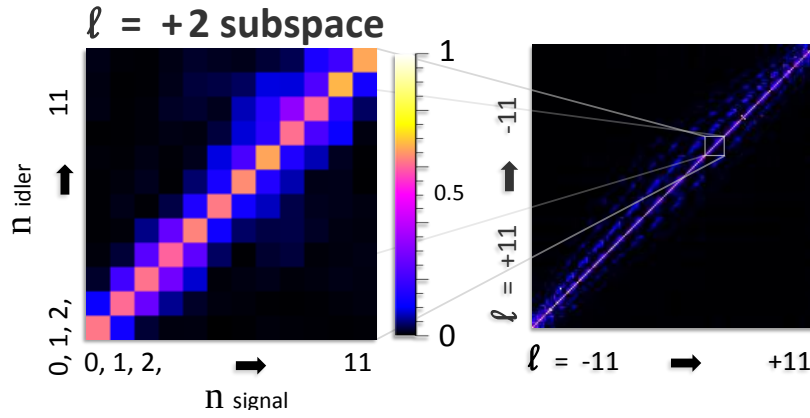


Figure 3.5: **a:** Normalized coincidence rate of different modes (with logarithmic scale), depending on the two mode-numbers (full-field bandwidth). It shows that the probability of higher-order modes drop exponentially, which is due to the creation process in the crystal [91] as well as due to inefficient coupling of higher-order modes. The absolute count rate was 105,500 photon pairs per second for a pump power of 60 mW. To be precise, this is the summed count rate of all 186 modes, not taking into account the inefficiencies of the detectors or imperfect coupling into SMFs. **b:** Weighted correlations between different modes in z basis. Due to different probabilities of different modes, in these pictures we weigh every correlation with the probability of the modes involved. That means, $\langle i|j \rangle_{\text{weighted}} = N \frac{\langle i|j \rangle_{\text{measured}}}{\sqrt{\langle i|i \rangle \langle j|j \rangle}}$ where i and j stand for different two-photon modes, and N is a normalization constant and $\langle i|j \rangle$ stand for the coincidence count rate when the first photon is projected into mode i and the second photon is projected into mode j . **b** (Left): The correlation of modes with $\ell = 2$ is shown, and reveals good correlation of modes with the same number of radial nodes. (Right): All correlations in the z basis are visualized.

The second quantum number n corresponds to the radial nodes in the intensity profile. Only lately this second degree of freedom has been analyzed theoretically in a quantum mechanical framework [16–18, 91–93]. In the down-conversion process the angular momentum of the photons is conserved, therefore this degree of freedom is anticorrelated. For the radial quantum number n the situation is more complicated. The full down-conversion process concerning the

3 Quantum Entanglement in 100 dimensions

correlations for the radial quantum number has been analyzed in detail [91] and quasiperfect correlations have been found for specific situations. Recently, these quasiperfect correlations have been demonstrated experimentally [94]. The state we expect from can be written as perfectly (anti-) correlated pure state $|\psi\rangle = \sum_{n=0}^{\infty} \sum_{\ell=-\infty}^{\infty} a_{n,\ell} |LG_{n,\ell}\rangle |LG_{n,-\ell}\rangle$, with ℓ and n dependent coefficients a . In the derivation of the bounds in equation (3.4) we restricted the states to be perfectly correlated. This means we assume a physical property of our input state, namely perfect (anti-)correlation of the modes. Small deviations from this assumption (which we have observed in the experiment) have been analyzed numerically and we found that they only reduce our observed W . Thus the application of the sufficient criterion in inequation (3.4) is justified in our experiment.

The experimental analysis of the LG modes of the photon pair produced is done by a holographic mode transformation using a spatial light modulator (SLM). With that we can transform any desired mode to a Gauss mode. By using a single-mode fiber (SMF), we filter only for Gauss modes and thereby project the quantum state into the desired mode [3]. The setup and exemplary LG modes are shown in figure 3.4.

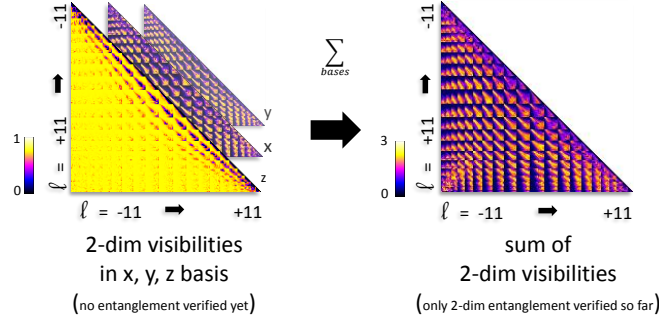


Figure 3.6: (Left) The visibilities of all of the two-dimensional subsets in all of the three bases are shown. The z visibility is usually larger, because non-maximal entanglement (figure 3.5A) reduces visibility in the x and y basis only. When we sum up the three visibilities (Right), we can see that some subsets are more entangled than others. Every subspace with a value larger than 1 is two-dimensional entangled; this is true for most of the 17,000 subspaces. Modes with similar count rates have high visibilities, whereas modes with a very different count rate have very low visibilities in the x and y bases. To reveal information about the global, high-dimensional entanglement, we have to sum up all visibilities of all subsets and calculate quantity W in equation (3.3).

In our experiment we analyze the correlations of 186 modes of two photons (figure 3.5). The number of modes, 186 in our case, corresponds to D in equation (3.4). We use LG modes with an angular quantum number up to $\ell = 11$, and a radial quantum number up to $n = 13$. To calculate the quantity in equation (3.3), we need to measure in every two-dimensional subspace (there are $\frac{186 \times 185}{2} = 17,205$ two-dimensional subspaces) the visibility in x , y , and z basis, which corresponds to 3×4 measurements per subspace. Altogether this results in 200,000 measurements (with 750 million detected photon pairs). For comparison, if we had performed a full state tomography, we would have needed to perform more than 1 billion measurements.

3 Quantum Entanglement in 100 dimensions

When we sum up all of our measured visibilities (see figure 3.6) according to equation (3.3), we find

$$W_{D=186} = 35,529 \pm 6 \quad (3.6)$$

which corresponds to at least 100-dimensional entanglement according to inequality (3.4) (101-dimensional: $W > 35,619$; 100-dimensional: $W > 35,433$; 99-dimensional: $W > 35,247$). The confidence interval corresponds to one standard deviation due to the statistical uncertainty. It has been calculated using Monte Carlo simulation. The detected photon numbers are assumed to be Poisson distributed, which leads to asymmetric distribution especially for low count rates. Analytical treatment of error propagation for such a large number of measurements was not practical. Therefore, all confidence intervals have been calculated using Monte Carlo simulations. The statistical uncertainty is very small because is calculated from 200,000 measurements with a large total count rate. The detailed measurement results and the calculation of (3.6) can be seen in figure 3.6 and 3.7.

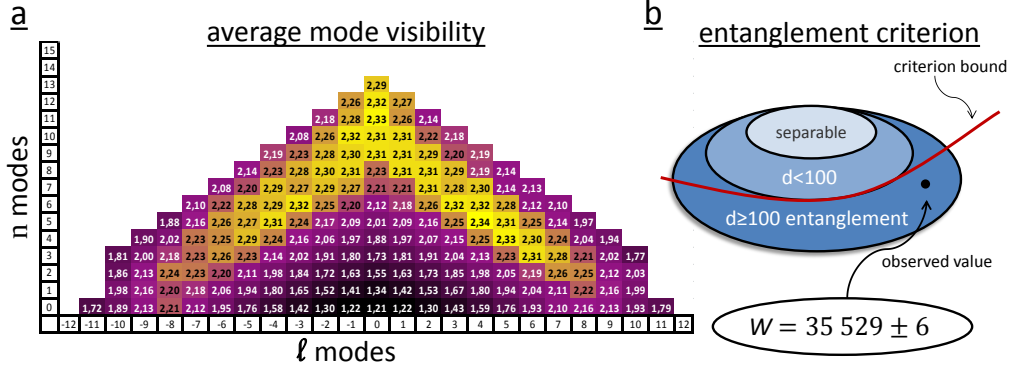


Figure 3.7: (A) The average sum of visibilities (in x, y, and z basis) of a specific mode with all other modes is shown. The observable structure originates from non-maximally entanglement due to different count rates for different modes (figure 3.5A). The bright regions in the center are modes with a similar probability. The central low-order modes (such as the Gauss mode) have the highest probability, therefore the lowest average visibility. Precisely, the Gauss mode has an average sum of visibilities of 1,21 (which mainly results from the visibilities in z basis). A maximally entangled high-dimensional state would have a summed visibility of 3 for every mode pair. The structure shows that different modes contribute differently to the entanglement criterion. (B) To reveal information about the global, high-dimensional entanglement, we have to calculate the value for the quantity W in equation (3.3). This is done by summing up all visibilities of all subsets. Here, the concept and result of the entanglement dimensionality criterion is visualized. With our criterion in equation (3.4), quantum states are divided into two parts: those with entanglement dimensionality smaller than 100 and those with a larger one (red line). We have observed a value that lies in the lower part, thus we have verified 100-dimensional entanglement.

3.3.1 Considered Modes vs. Entanglement Dimensionality

The quantity W in equation (3.3) corresponds to measurements of all two-dimensional subspaces in a $(D \times D)$ -dimensional quantum state. It can be seen in figure 3.7A that some modes contribute more to the quantity than others, thus we can try to find a smaller optimal set of modes that shows the higher-dimensional entanglement. We find that by removing 19 modes (that means, not taking into account all two-dimensional subspace measurements with them), we can find at least 103-dimensional entanglement (figure 3.8).

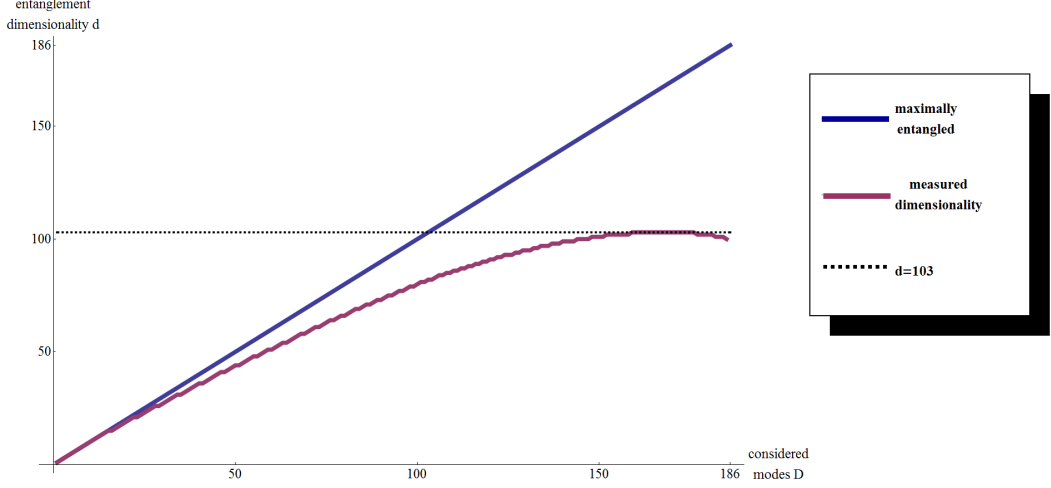


Figure 3.8: The entanglement dimensionality d depends on the number of modes considered for W , as described in inequality (3.4) in the main text. We measured all two-dimensional subspace correlations in x , y , and z bases for 186 different modes. If the state would be maximally entangled, one could extract as much entanglement dimensionality as one considers modes (blue line). However, as it can be seen in figure 3.7A, there are modes which contribute stronger to W (yellow region), and some modes that contribute less to the witness (for instance the Gaussian mode). One can remove certain modes (i.e., not considering all two-dimensional subspace measurement that contains these specific modes). We find a maximal detectable entanglement dimensionality d , if we only consider the 167 strongest contributing modes (red curve). This leads to 103-dimensional entanglement (black dashed line). We also observe a set of 15 modes which are entangled in 15 dimensions, which could be significant in special protocols in quantum communication.

To explain this behaviour more, we give a simple example: Consider the four-dimensionally entangled pure state

$$|\psi\rangle = N (0.5|0, 0\rangle + 0.07|1, 1\rangle + 0.01|2, 2\rangle + 0.01|3, 3\rangle) \quad (3.7)$$

where N is the normalization, we can calculate the subspace visibilities. The sum of the x , y , and z visibilities in every subspace ($SV = V_x + V_y + V_z$):

$$SV^{0,1} = 1.55, SV^{0,2} = 1.08, SV^{0,3} = 1.08, SV^{1,2} = 1.56, SV^{1,3} = 1.56, SV^{2,3} = 3.$$

3 Quantum Entanglement in 100 dimensions

Calculating the sum of all visibilities (according to equation (3.3)) gives $W = 9.723$. Comparing with the inequality from (3.4) in the main text, using $D = 4$, we can confirm a two-dimensional entanglement (bounds are 6, 10, and 14 for two-, three-, and four-dimensional entanglement, respectively). If we only consider modes 1, 2, and 3 (and do not consider mode 0), we get $W = 6.12$, which confirms a three-dimensional entanglement (bounds are 3 and 6 for two- and three-dimensional entanglement, respectively). This example shows how considering a smaller number of modes can verify a higher-dimensional entanglement with our method.

3.3.2 Effect of Deviation from Perfect Correlation

For the derivation of the bounds in (3.4), we assumed perfect correlations between all involved modes. This is physically motivated. The first quantum number corresponds to the angular momentum of light, which is known to be conserved in the down-conversion process in the paraxial approximation [3]. The second quantum number is the radial momentum, which are correlated as well, as can be seen in our experimental data in figure 3.5B.

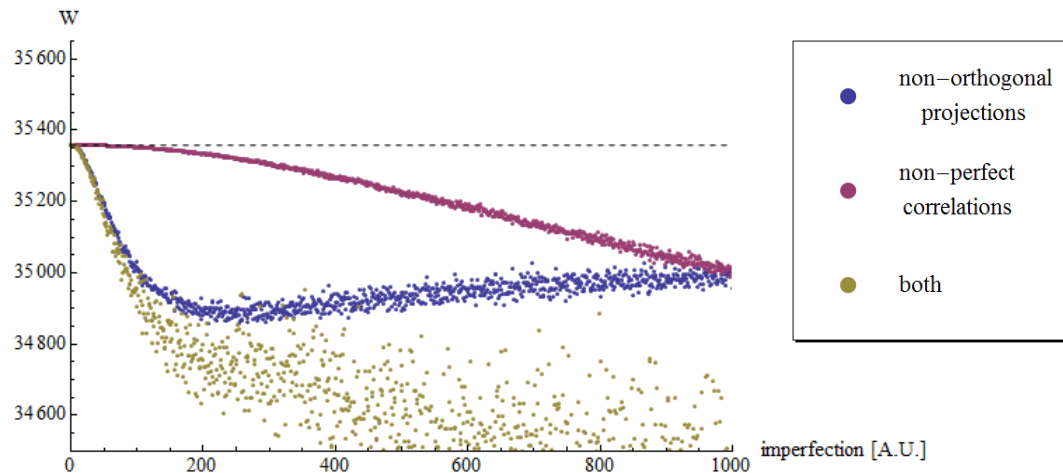


Figure 3.9: A non-maximally entangled state with $D = 186$ dimensions is considered, similar to the state we expect from our experiment. It is considered how nonorthogonal projections and other measurement-induced errors (blue), nonperfect correlations (red), and both effects simultaneously (yellow) influence the value of the quantity W . For each type, 1,000 cases are calculated. The imperfections are introduced randomly, in each step the introduced imperfections increase. The dashed black line shows the value calculated without imperfections. It can be seen that any introduction of nonperfect correlations or nonorthogonal projections only decreases W . These results guarantee that the experimentally observed deviation from the perfectly correlated state cannot artificially increase the observed dimensionality (in contrast, it decreases the quantity W thus the observed Schmidt number).

Deviations from this assumption have been analyzed numerically, see figure 3.9. We have analyzed how nonperfect correlated states or nonorthogonal projections influence our quantity W (3.3). We have found that for small deviations such as observed in our experiment, W can only decrease, thus the resulting bounds still hold. Therefore, the application of our

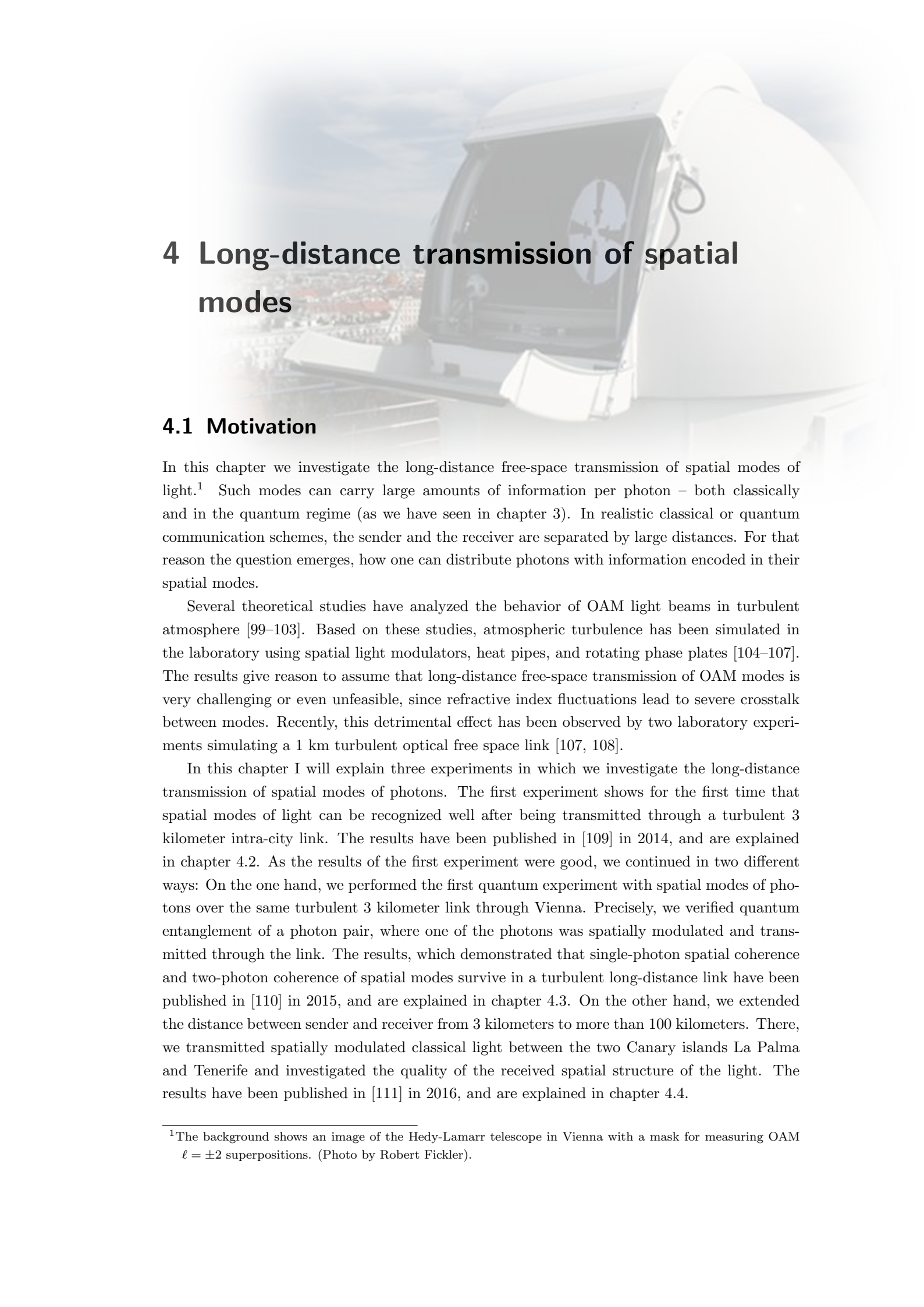
entanglement dimensionality criterion is justified.

3.4 Discussion

One way to bring this in relation with other photonic and multipartite entanglement experiments is the following. The dimension of the entangled Hilbert-space scales with $\dim = d^N$, where d stands for the entangled dimensions and N is the number of involved parties. Our experiment shows an entangled Hilbert space dimension of at least $\dim = (103 \times 103) \approx 2^{13.4}$ that is larger than the largest entangled photonic Hilbert space reported so far (with $\dim = 2^{10}$) [95, 96]. Interestingly, it is of similar magnitude as that of the largest quantum systems with multipartite entanglement measured so far, such as 14-qubit ion entanglement with $\dim = 2^{14}$ [60].

Our results show that we can experimentally access a quantum state of two photons which is at least (103×103) -dimensionally entangled. This was possible by developing a new method to analyze efficiently and in an experimentally practical way quantum states with very high dimensions. The entanglement dimensionality witness used in this work has further been improved into a state-independent form without any physical assumptions in [97], which can also be applied to multi-photon high-dimensional experiments [98].

Such high-dimensional entanglement offers a great potential for quantum information applications. There are situations where two-dimensional entanglement is no longer sufficient but high-dimensional entangled systems are able to perform the task [65]. In realistic quantum cryptography schemes, for example where noisy environment or manipulated random number generators lead to a breakdown of the system for low-dimensional entangled states, high-dimensional entanglement sustains the security [63–65]. The experimental setups as presented here are suitable for such tasks. A big open question is how photons encoded in such spatial modes can be transmitted between distant parties – a task necessary for any classical or quantum communication protocol. We are going to investigate that question in the next chapter.



4 Long-distance transmission of spatial modes

4.1 Motivation

In this chapter we investigate the long-distance free-space transmission of spatial modes of light.¹ Such modes can carry large amounts of information per photon – both classically and in the quantum regime (as we have seen in chapter 3). In realistic classical or quantum communication schemes, the sender and the receiver are separated by large distances. For that reason the question emerges, how one can distribute photons with information encoded in their spatial modes.

Several theoretical studies have analyzed the behavior of OAM light beams in turbulent atmosphere [99–103]. Based on these studies, atmospheric turbulence has been simulated in the laboratory using spatial light modulators, heat pipes, and rotating phase plates [104–107]. The results give reason to assume that long-distance free-space transmission of OAM modes is very challenging or even unfeasible, since refractive index fluctuations lead to severe crosstalk between modes. Recently, this detrimental effect has been observed by two laboratory experiments simulating a 1 km turbulent optical free space link [107, 108].

In this chapter I will explain three experiments in which we investigate the long-distance transmission of spatial modes of photons. The first experiment shows for the first time that spatial modes of light can be recognized well after being transmitted through a turbulent 3 kilometer intra-city link. The results have been published in [109] in 2014, and are explained in chapter 4.2. As the results of the first experiment were good, we continued in two different ways: On the one hand, we performed the first quantum experiment with spatial modes of photons over the same turbulent 3 kilometer link through Vienna. Precisely, we verified quantum entanglement of a photon pair, where one of the photons was spatially modulated and transmitted through the link. The results, which demonstrated that single-photon spatial coherence and two-photon coherence of spatial modes survive in a turbulent long-distance link have been published in [110] in 2015, and are explained in chapter 4.3. On the other hand, we extended the distance between sender and receiver from 3 kilometers to more than 100 kilometers. There, we transmitted spatially modulated classical light between the two Canary islands La Palma and Tenerife and investigated the quality of the received spatial structure of the light. The results have been published in [111] in 2016, and are explained in chapter 4.4.

¹The background shows an image of the Hedy-Lamarr telescope in Vienna with a mask for measuring OAM $\ell = \pm 2$ superpositions. (Photo by Robert Fickler).

Several other groups have investigated similar questions in recent years. In particular, in 2012, a classical communication experiment with twisted radio waves was performed over 420 meters in Venice [112]. In 2014, single photons carrying OAM in the visible frequency range were transmitted over an indoor 210 meters in a QKD experiment in a large hall in Padua [113]. In 2015, one experiment in Erlangen tested classical transmission and cross-talk of OAM beams over 1.6 kilometers [114]. Finally in 2016, a high-speed classical communication experiment using OAM mode multiplexing was performed over 120 meters of free-space in Los Angeles, transmitting 400 GBit/sec [115].

4.2 Classical transmission over an intra-city link

The content of this chapter is based on the publication "Communication with spatially modulated light through turbulent air across Vienna" [109]².

The theoretical and lab-simulation studies mentioned above employed coherent mode detection. Different OAM beams are generated from Gaussian beams using holographic transformations at the sender and were then transmitted through simulated atmosphere. At the receiver, the transmitted modes are transformed to Gaussian beams in order to analyze the quality of the OAM modes after free-space propagation. However, such transformations [3, 116, 117] rely on axis-dependent decomposition of modes and are extremely sensitive to angle of arriving (AOA) fluctuations and beam wander. As the effect of atmospheric turbulence on the beam propagation can be decomposed into different orders of Zernike polynomials [118], the difficulty in using such techniques can be explained rather intuitively. Beam wander (lateral tilts) is the first order contribution of the atmosphere, which influences such phase measurement based detection schemes significantly. Additionally, higher order contributions such as defocusing, astigmatism or coma further reduce the mode transformation quality and efficiency, for instance due to inefficient identification of a defocused Gaussian mode.

In the experiment presented here, OAM superposition modes of light are transmitted over a 3 km intra-city link in Vienna under strong turbulence conditions. We employ an incoherent detection scheme by directly observing the unambiguous mode intensity patterns on a screen with the help of an adaptive pattern recognition algorithm. This method avoids coherent phase-dependent measurements for identifying the transmitted OAM modes, therefore is not affected by the atmospheric contribution described above. In particular, the disadvantageous effect of beam wander is reduced significantly as the method is not axis-dependent. We transmitted 16 different mode superpositions ($\ell=0, \pm 1, \dots, \pm 15$) and could distinguish them with an average error of only $\approx 1.7\%$. Additionally, we analyzed the atmospheric effect on the relative phase of these superpositions, which is a crucial property for verifying quantum entanglement of OAM modes in future experiments. We found that the relative phase is only slightly affected by turbulence which favors the use of petal patterns in the experiment. Petal patterns we call the intensity pattern of superpositions of modes with $+\ell$ and $-\ell$. It leads to 2ℓ intensity minima and maxima symmetrically arranged in a ring. That finding motivated us to perform two follow-up

²Mario Krenn, Robert Fickler, Matthias Fink, Johannes Handsteiner, Mehul Malik, Thomas Scheidl, Rupert Ursin, Anton Zeilinger, Communication with spatially modulated light through turbulent air across Vienna. *New Journal of Physics* **16**, 113028 (2014).

4 Long-distance transmission of spatial modes

experiments: The first one exploited the stability of relative phases to verify long-distance OAM entanglement using a polarization-OAM hybrid entangled state. It is described in chapter 4.3. In the second follow-up experiment, we extended the distance between sender and receiver to 143 kilometers, which is described in 4.4.

We have chosen OAM superposition modes due to the fact that well developed methods exist to detect quantum entanglement in future experiments. However, this approach does not only work for OAM superposition modes, but for every spatial mode structure with unambiguously distinguishable intensity patterns. The highly symmetric petal patterns of OAM superpositions seem to be advantageous in turbulent conditions in order to correctly identify the modes with a pattern recognition algorithm. It might be interesting to analyze whether different spatial mode structures (such as Hermite-Gaussian beams, self-healing Bessel beams [119] or more complex structures such as Ince-Gaussian modes [19]) are identifiable in a more robust way than Laguerre-Gauss modes.

4.2.1 Experimental setup

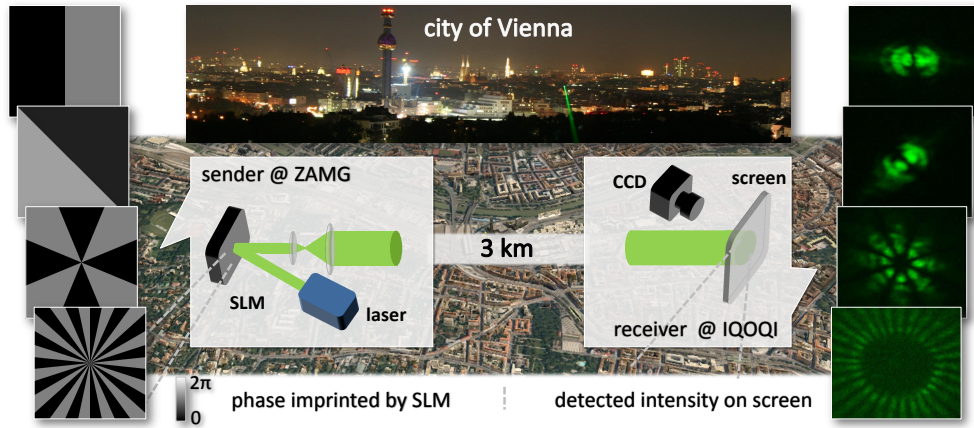


Figure 4.1: Sketch of the experimental setup. The 3 kilometer free-space experiment was performed in the city of Vienna, from ZAMG (*Zentralanstalt für Meteorologie und Geodynamik, Central Institute for Meteorology and Geodynamics*) to our institute IQOQI. Top: Picture of an alignment laser beam from IQOQI to ZAMG (the green laser can be seen roughly at the center of the image), captured at ZAMG. Left: The sender modulates a 532nm laser with an SLM (Spatial Light Modulator). Different phase holograms that modulate the beam are shown; they correspond to superpositions of OAM modes (from top to bottom) with $\ell = \pm 1$, $\ell = \pm 1$ rotated, $\ell = \pm 4$ and $\ell = \pm 15$. Right: At the receiver we observe the transmitted modes and record them with a CCD camera. The images correspond to the modulated phases on the left. The size of the inner most intensity ring varies from approximately 12cm for $|LG_{\pm 1}\rangle$ to 72cm for $|LG_{\pm 15}\rangle$. By analyzing the observed images, we characterize the atmospheric stability of the OAM modes and use them for transmitting real information. (Geographic pictures taken from Google Earth, 2014 ©Google, Cnes/Spot Image, DigitalGlobe)

The experimental setup for sending and receiving OAM modes over the city of Vienna can be

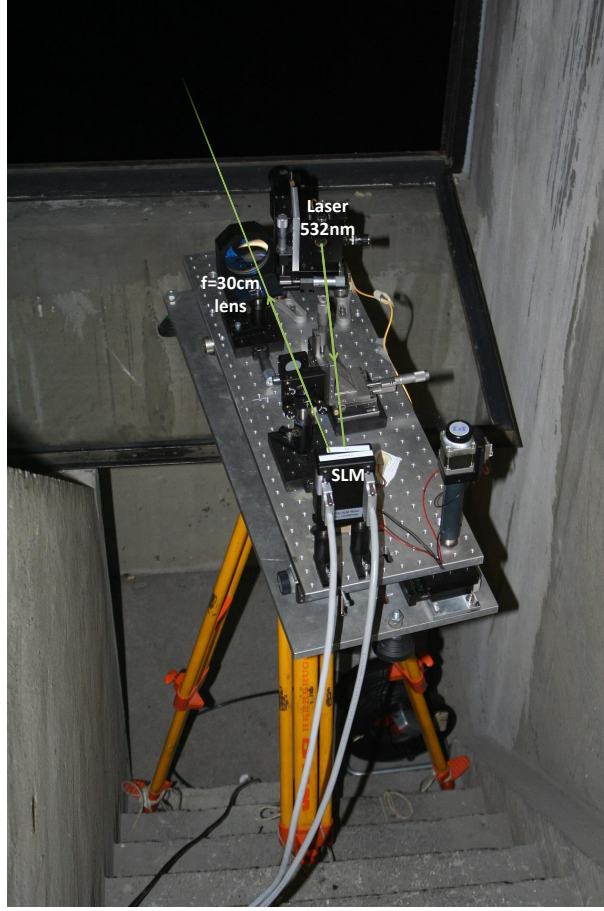


Figure 4.2: Picture of the sending telescope, with beam path indicated in green, and key components described in white.

seen in figure 4.1. The sender was located in a $\approx 35\text{m}$ high radar tower of ZAMG (*Zentralanstalt für Meteorologie und Geodynamik, Central Institute for Meteorology and Geodynamics*). The receiver was on the rooftop of our institute building 3040 meters away. At the sender (see picture in figure 4.2 and 4.3), we had a 20mW laser with a wavelength of 532nm. The light from the laser was modulated by a Spatial Light Modulator (SLM) which impressed the phase information of the OAM modes onto the beam. Then it was expanded by a telescope to a diameter of approximately 6 cm and sent with a high-quality $f=30\text{cm}$ lens to the receiver. The received mode intensity profiles were observed on a screen, and recorded with a CCD camera.

We characterized the turbulence of the atmosphere in the following way: The atmospheric turbulence can be decomposed into cells with similar pressure, thus similar refractive index. Those *Fried cells* are flowing across the propagating path and randomly deflect the centroid of the beam in different directions. If the light beam is observed on a screen it *jumps* around on a time scale of 1 kHz. However, when averaged over time, the deviation of the intensity's centroid position appears small. The magnitude of the short-term beam wander depends on the refractive index structure parameter C_n^2 and the path length L . It is characterized by the root-mean-square of the beam displacement from the time-averaged center. We measure the

4 Long-distance transmission of spatial modes



Figure 4.3: Image of the sending station at the radio tower at ZAMG. The upper image is taken during the day, the lower image is recorded during the night. In the lower image, there are two lasers visible: A faint green laser beam which is sent from the ZAMG tower to the receiver (difficult to see, on the left of the strong green laser), and a strong green laser beam which is used as alignment laser from the IQOQI Vienna building to the ZAMG tower. The strong laser can also be seen in figure 4.1 in the top image.

root-mean-square of Gaussian beam size with images of 1/2000sec and 1/4000sec integration time, and with images of 20 seconds integration time. From these values we calculated a Fried parameter of $r_0=1.4\text{cm}$ and an atmospheric structure constant of $C_n^2 = 7.7 \cdot 10^{-15} \text{m}^{-\frac{2}{3}}$ using the Kolmogorov theory of atmospheric turbulence [120–122]. These parameters correspond to strong turbulence conditions [107, 108].

In the experiment we sent light in superpositions of higher-order OAM modes. The complete state can be denoted by

$$|\text{LG}_{\pm\ell}^\alpha\rangle = \frac{1}{\sqrt{2}} (|\text{LG}_{+\ell}\rangle + e^{i\alpha}|\text{LG}_{-\ell}\rangle), \quad (4.1)$$

where α denotes the relative phase between the two modes, which corresponds to a rotation of the phase and intensity structure. The LG_ℓ are Laguerre-Gauss modes (2.19) with the radial quantum number $n = 0$. The transverse phase structure of an OAM-superposition $|\text{LG}_{\pm\ell}\rangle$

is radially symmetric and has 2ℓ phase jumps of π in the azimuthal direction. Its intensity distribution shows 2ℓ maxima and 2ℓ minima arranged in a ring (figure 4.1).

The size of the detected modes varied between 12cm for $|\text{LG}_{\pm 1}\rangle$ to 72cm for $|\text{LG}_{\pm 15}\rangle$, where the final beam size variation is mainly due to diffraction. The size of the intensity structure of our modes scales linearly with ℓ , as it is expected for holographic generation without intensity shaping [29]. The size can be reduced significantly by applying intensity shaping on the SLM or using non-diffractive beams such as Bessel beams. In our experiment however, the beam size was not an issue.

4.2.2 Mode detection

Artificial neural network

To analyze the intensity structure of the received modes, we use an adaptive pattern recognition algorithm in form of an artificial neural network [123, 124]. In the *training* or initialization phase, the algorithm receives a number of recorded images in order to autonomously learn how to recognize the different patterns. After this initialization, the algorithm is ready to analyze the real data in form of images. The working principle is explained with a simplified example in figure 4.4. There, as an example, a network with a chain of 50 neurons is initialized (Figure

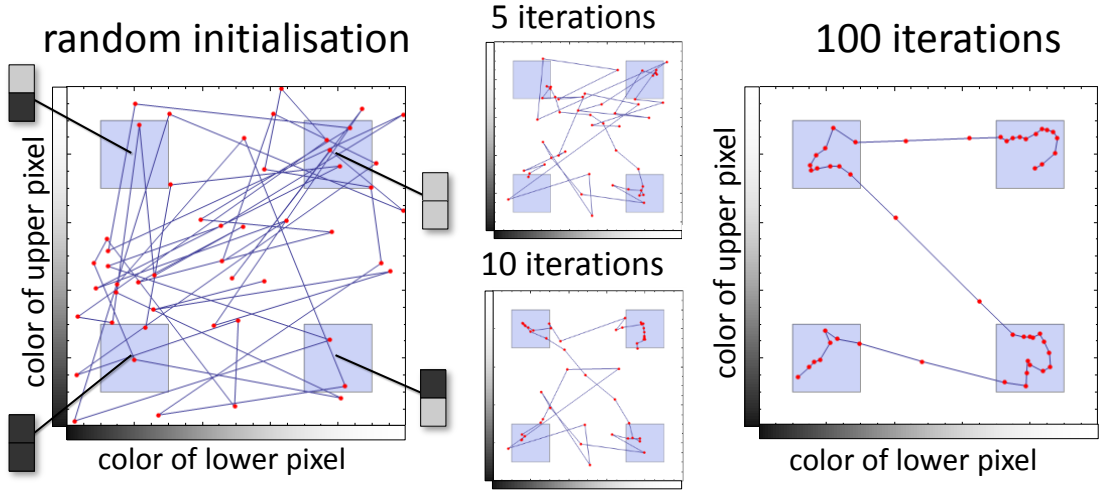


Figure 4.4: Working principle of the artificial neuronal network (ANN) described by an example of four different grey-scale images with only two pixels each.

4.4 left). The neurons (red dots) are connected to their neighbors, as indicated by the blue lines. Every neuron is assigned a random position in 2-dimensional space which corresponds to their grey-value (the color of the upper pixel corresponds to a position at the y-dimension; the lower one corresponds to the x-axis). In each subsequent step, the ANN receives one out of four possible 2-pixel images (dark-dark, dark-bright, bright-dark, bright-bright), which as well corresponds to a coordinate in the 2-dimensional space. Due to *virtual turbulence*, the color of the images might be changed slightly, resulting in a 2-dimensional region (indicated in blue) rather than a single position. For each input image, the position of the winner neuron (the neuron closest to the input image) is pulled strongly towards the input image. The positions of

4 Long-distance transmission of spatial modes

the winner neuron's neighbors are pulled towards the input picture as well - but less significantly. After a few iterations (Figure 4.4 middle), the initially random network organizes itself and finds the structure of the input in an unsupervised way (Figure 4.4 right). After the training phase, we can assign to each neuron the information to which image it corresponds (for example, the green neuron in the right picture belongs to the image dark-dark). For this we input perturbed images (for instance a dark-dark image), and assign the information about the image to the winner neuron (for example, to the green neuron). As soon as most neurons have been assigned a specific image information, the network can be used for identifying unknown images. To identify unknown images, we again calculate the winner neuron for each image, and get the image information assigned to the neuron. In the example above, 5 out of 50 neurons in the final network lie outside the blue area, and it is likely that they will never be a winner neuron, thus could be removed from the network (for computational speedup). In our experiment, the inputs were images with $720 \times 720 = 518.400$ pixels, thus the size of the virtual neuronal network space is 518.400-dimensional. In that space, the ANN autonomously categorizes the 16 input structures that describe our different OAM mode superpositions. As the neural network is trained with images involving atmosphere-induced disturbances, it is developing automatically a robust detection despite such effects.

Cross-correlations

In the first step, we investigate whether the characteristic mode patterns of different modes can be distinguished after free-space transmission. For that we analyze the crosstalk between the first 16 OAM mode superpositions $|\text{LG}_{\pm\ell}\rangle$ (with $\alpha=0$), from $\ell=0$ up to $\ell=15$, at the receiver. For each transmitted ℓ -value, we accumulated approximately 450 received mode-intensity images, which served as the input for our algorithm. As a result, we obtained the detected ℓ -value of the received mode. By comparing the prepared and measured ℓ -values, we can calculate the crosstalk matrix between the different OAM modes, which is shown in figure 4.5A. For superpositions of $|\text{LG}_0\rangle$ up to $|\text{LG}_{\pm 15}\rangle$ we find a good distinguishability with an average error of 1.7%. The error is defined as the ratio between wrongly detected modes and all detected modes.

4.2.3 Data transmission encoded in OAM

To illustrate the quality of the received modes, we used these 16 different states for encoding two greyscale images with 8 and 16 different greyscale values (corresponding to 3bits and 4bits per pixel, respectively). Each transmitted mode carried the information of one pixel of the image. The mode $|\text{LG}_0\rangle$ corresponds to black, while higher-order modes correspond to grey-values. The highest mode in the alphabet ($|\text{LG}_{\pm 7}\rangle$ for 3bits and $|\text{LG}_{\pm 15}\rangle$ for 4bits) corresponds to white. The maximum frame rate of the SLM is 60Hz, and for the camera we used 50Hz. As the SLM and the CCD camera were not synchronized, and the liquid crystal display of the SLM was slow due to the low environment temperature of $\approx 5\text{-}10^\circ\text{C}$, we displayed each mode for 10 SLM frames. In between two modes, the beam was deflected for 4 frames to distinguish between the subsequent modes. The transmission rate thus was 4 pixel/sec. The transmitted and decoded image can be seen in figure 4.5b. The errors in the decoded 3bit and 4bit images

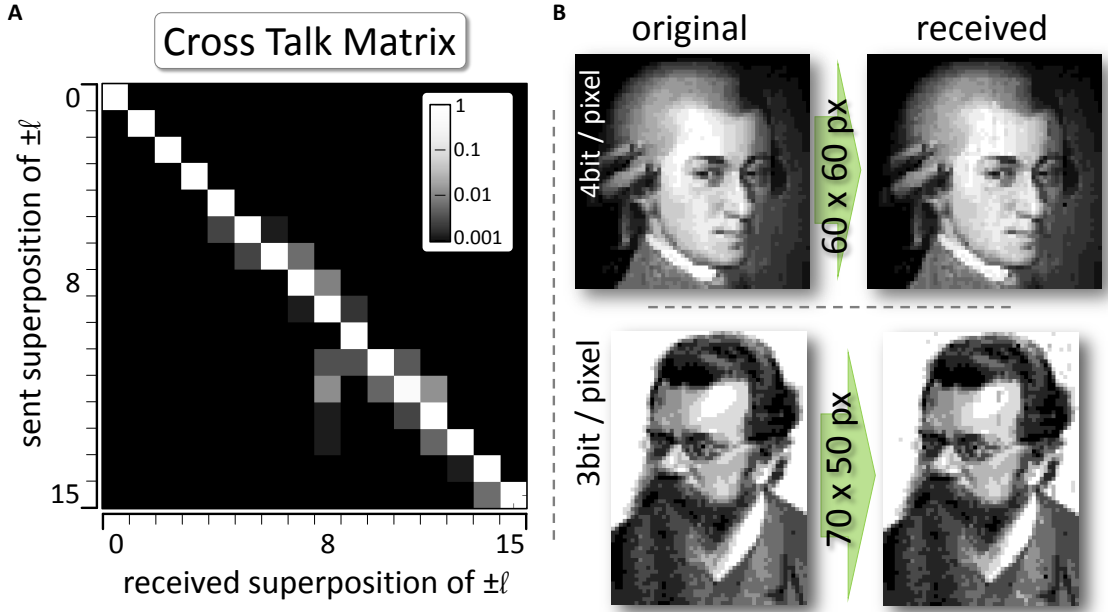


Figure 4.5: A) The crosstalk matrix for different OAM superposition settings (in logarithmic scale). The cross talk matrix shows the distinguishability of OAM mode superpositions from $|LG_0\rangle$ to $|LG_{\pm 15}\rangle$ (corresponding images in figure 4.1). We can distinguish the modes with an average error rate of 1.7% and a channel capacity of 3.89 bits. B) We transmit two greyscale images encoded in these OAM mode superpositions. The upper image (Wolfgang Amadeus Mozart) has 4 bits per pixel, which corresponds to 16 greyscale settings. As a result, the full available set of modes was used to encode this image. The received image has a bit-error-ratio of 1.2%. The lower image (Ludwig Boltzmann) has 3 bits per pixel, which required 8 different modes. The average error rate for this image is measured to be 0.8%.

are 0.8% and 1.2%, respectively. The bit-error is defined as the ratio between wrong bits in the decoded image and all bits of the images.

4.2.4 Relative phase stability

As mentioned above, the relative phase α of OAM superpositions is crucial for verifying quantum entanglement of OAM modes in future experiments. Hence, we analyzed in a second step the stability of α under atmospheric turbulence. By changing the relative phase, one can access the whole equator of the qubit Bloch sphere, thus has access to two different mutually unbiased bases (MUBs). Using two entangled photons, a high visibility in those two MUBs would be sufficient to verify entanglement [89]. For OAM modes, the relative phase rotates the intensity pattern of the mode, which we can directly observe.

We changed the relative phase in modes $|LG_{\pm 1}\rangle$ (see equation 4.1), $|LG_{\pm 2}\rangle$ with step sizes of 11.25° , and analyzed their distinguishability with the pattern recognition algorithm (figure 4.6). With this angle, the rotated modes could be distinguished with an error of 15.9% averaged for $|LG_{\pm 1}\rangle$ and $|LG_{\pm 2}\rangle$. When we omit every second measured phase angle, and analyze the

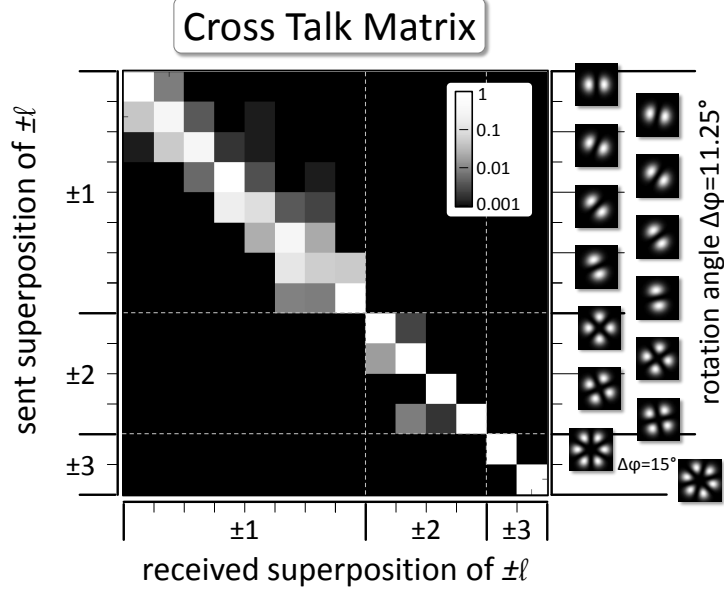


Figure 4.6: Cross Talk Matrix for rotated mode patterns (in logarithmic scale). We send OAM superpositions of $|\text{LG}_{\pm 1}\rangle$, $|\text{LG}_{\pm 2}\rangle$, and $|\text{LG}_{\pm 3}\rangle$ with different relative phases, which leads to rotated intensity patterns by $\Delta\phi$. The pattern recognition algorithm is able to distinguish them with an error-ratio of 0.7% for angles of 22.5° and an error of 15.9% for angles as small as 11.25° . The stability of relative phases is crucial for follow-up experiments on quantum entanglement of OAM over large distances.

relative phases in 22.5° steps, we find an average error of only 0.7%.

The smaller error in the case of a rotation angle 22.5° than for 11.25° is expected because the intensity patterns are rotated by twice the angle. Therefore they are easier to distinguish.

4.2.5 Remarks

In addition to the results from the artificial neural network, we also investigated long-term exposure images of the modes, and find that the mode quality was very good on average (figure 4.7). The small influence of the atmosphere on the relative phase α indicates the feasibility of quantum entanglement experiments with OAM over long distances. These (for us surprising) results have raised two immediate follow-up questions:

First, can quantum information be encoded and transmitted on single photons, and can entangled photon pairs be transmitted through such a distance? Such an experiment would answer whether single-photon spatial coherence as well as two-photon coherence is significantly distorted over large distances. We have investigated this question in a follow-up experiment explained in chapter 4.3.

Second, how far can information be transmitted with spatial modes of light when it passes through a turbulent atmosphere? Is the transmission of spatially modes of light over much larger distances (for instance with satellites, for which one needs to overcome an effective atmosphere of more than 10 kilometers) possible? We have performed an experiment over more than 140

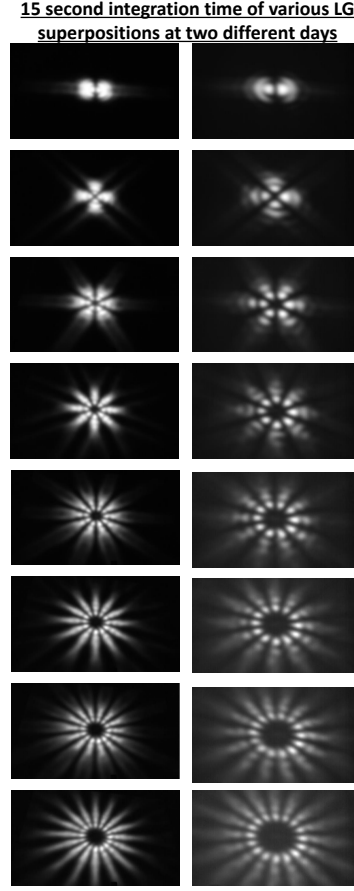


Figure 4.7: Long-time exposure of LG superpositions with various ℓ . Each image is integrated over 15 seconds, the left and right column are recorded on two different days. The first row has $\ell = \pm 1$, and increases by one per row, until the last row with $\ell = \pm 8$.

kilometers, which is explained in 4.4.

4.3 Quantum Entanglement over an intra-city link

The content of this chapter is based on the publication *"Twisted photon entanglement through turbulent air across Vienna"* [110]³.

Long-distance quantum entanglement with photons opens up the possibility to test fundamental properties of quantum physics in regimes not accessible in lab scale experiments. It can be used for quantum communication between widely separated parties, and it is the basis of quantum repeaters as nodes in a global quantum network. As the polarization of photons is easily controllable and resistant against atmospheric turbulence, it has been successfully used in a variety of different long-distance quantum experiments [125–128]. However, the polariza-

³Mario Krenn, Johannes Handsteiner, Matthias Fink, Robert Fickler, Anton Zeilinger, Twisted photon entanglement through turbulent air across Vienna. *PNAS* **112**, 14197-14201 (2015).

4 Long-distance transmission of spatial modes

tion of photons resides in a two-dimensional state-space, restricting the complexity of entangled states both for certain quantum communication tasks and for fundamental tests.

In contrast to polarization, the spatial modes of photons have an unbounded state-space, thus they can carry larger amount of information per photon. However, the possibility of more complex entangled quantum states poses a substantial challenge due to the negative influence of atmospheric turbulence on such modes. As mentioned before, several theoretical [101, 129–134] and lab-scale [104, 106, 135] studies investigated the effect of turbulence on entanglement encoded in the OAM of photons. The only OAM quantum experiment carried out beyond the lab-scale was located in a large hall to minimize the disturbing effects of turbulence [113]. Other than that, no experiment at the quantum level has been performed in a long-distance turbulent real-world environment, and quantum entanglement has not yet been demonstrated beyond the lab-scale with photons carrying OAM.

In the experiment explained in chapter 4.2 we found indications that the phase of OAM superpositions is well conserved during the transmission, hinting that the distribution of quantum entanglement encoded in OAM might be possible. In this section we discuss the follow-up experiment which deals with single-photon transmissions and distribution of entanglement in the OAM degree of freedom over the same 3 kilometer intracity link. With that, we confirm the indication of the first experiment mentioned above: We show that quantum entanglement distribution with spatial modes is possible over a turbulent intra-city link.

4.3.1 Experimental setup

The experimental setup can be divided into four main parts, see figure 4.8: the source of polarization entangled photon pairs, the transfer of one of those photons from polarization to the OAM degree of freedom, Alice’s polarization analysis and Bob’s OAM measurement after transmission. The sender (Alice) and the receiver (Bob) are at different physical locations 3 kilometers apart. The sender is located in a ≈ 35 meter high radar tower of Zentralanstalt für Meteorologie und Geodynamik (ZAMG - Central Institute for meteorology and geodynamics). There we use a high-fidelity high-brightness polarization entanglement source [136, 137] with an uncorrected average visibility of $\approx 97.5\%$. Photon A is unchanged, whereas photon B’s polarization state is transferred interferometrically to an OAM state [44]. After the transfer, the generated hybrid-entangled quantum state can be written as

$$|\psi\rangle = \frac{1}{\sqrt{2}} (|H\rangle_A |LG_{+\ell}\rangle_B + |V\rangle_A |LG_{-\ell}\rangle_B). \quad (4.2)$$

To ensure that photon B is transferred and sent before photon A is measured, we delay photon A by a 30 meter fiber. It ensures that the detection of the two entangled photons is space-like separated: Any signal from one measurement to the other would need to propagate faster than the speed of light to influence the result. Afterwards the polarization of photon A is measured with a two-output polarization analyzing station. Each detection event is time-stamped and recorded with a time tagging module (*Austrian Institute Of Technology TTM 8000*). The OAM-encoded photon B is magnified to a Gaussian beam waist of 11 millimeters and send through turbulent atmosphere with a high quality lens (best-form f/4 doublet lens, with low spherical aberrations) using a focal length of $f=30\text{cm}$ to the receiver 3 kilometers away.

4 Long-distance transmission of spatial modes

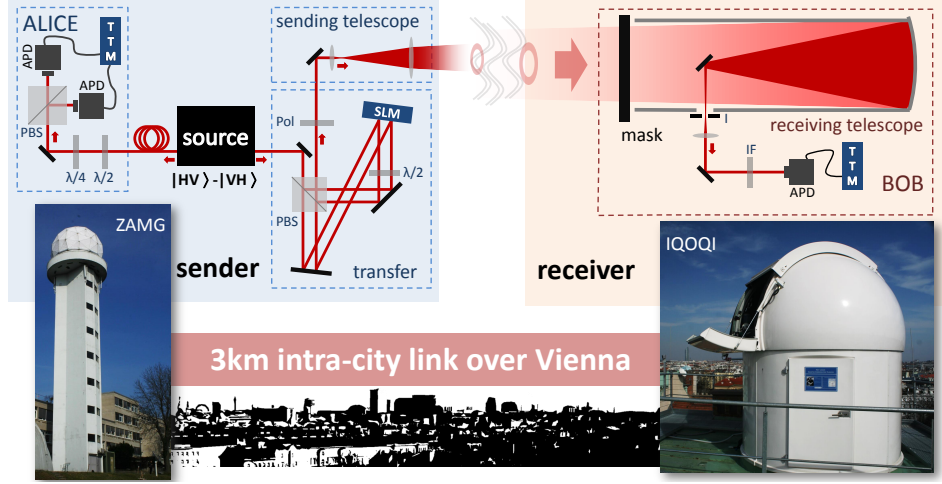


Figure 4.8: Sketch of the experimental setup. The experiment takes place at two locations separated by 3 kilometers. The sender is located in a radar tower of ZAMG (*Zentralanstalt für Meteorologie und Geodynamik*), the receiver is at the rooftop of our institute IQOQI (*Institute for Quantum Optics and Quantum Information*). Left: At the sender, we have a high-fidelity Sagnac-type polarization entanglement source. While photon A remains in the polarization degree of freedom, photon B is transferred to OAM, using an interferometric scheme [44, 138]: In it, the photon's path is separated according to its polarization at a polarizing beam splitter (PBS) and transformed to an OAM value depending on its path using a spatial light modulator (SLM, Hamamatsu LCOS-SLM). After recombination of the paths, the transfer is completed by deleting the polarization information with a polarizer (Pol). Subsequently, the photon wave front is expanded and sent to the transceiver with a high-quality lens. Meanwhile photon A of the entangled pair is delayed in a 30 meter fiber to ensure the transfer and sending of photon B before photon A is detected. After the fiber photon A is measured using a half-wave plate ($\lambda/2$) or a quarter-wave plate ($\lambda/4$) - depending on the basis in question -, a PBS and two avalanche photon detectors (APDs). The detection times of the photons are recorded with a time tagging module (TTM). Right: At the receiver, the transmitted photons are collected by a Newton-type telescope with a primary mirror of 37 cm diameter. In front of the primary mirror, opaque masks with symmetric slit patterns are used to perform mode measurements (figure 4.10). An iris (I) and a 3nm band pass filter (IF) were used to minimize background light. The photons are detected with an APD, and time tagged with a TTM. Coincidences are then extracted by comparing the time tagging information from both locations.

At the receiver on the rooftop of our institute IQOQI Vienna, we use a Newton-type telescope with a primary mirror of 37 cm diameter and a focal length of $f=1.2\text{m}$. In front of the primary mirror, we use an absorptive mask with a transparent, symmetric slit pattern to measure the modes (figure 4.9 and 4.10). The technique [44] allows us to measure visibilities in OAM-superposition bases, which is sufficient to verify entanglement. The masks are 40 cm in diameter and have a slit opening angle of 16° and 5.6° (for $\ell=1$ and $\ell=2$, respectively). The transmitted light is then detected on an APD with an active area of $500\mu\text{m}$ diameter. Similarly to the detection of the polarized partner photons, the arrival times are time tagged with a sec-

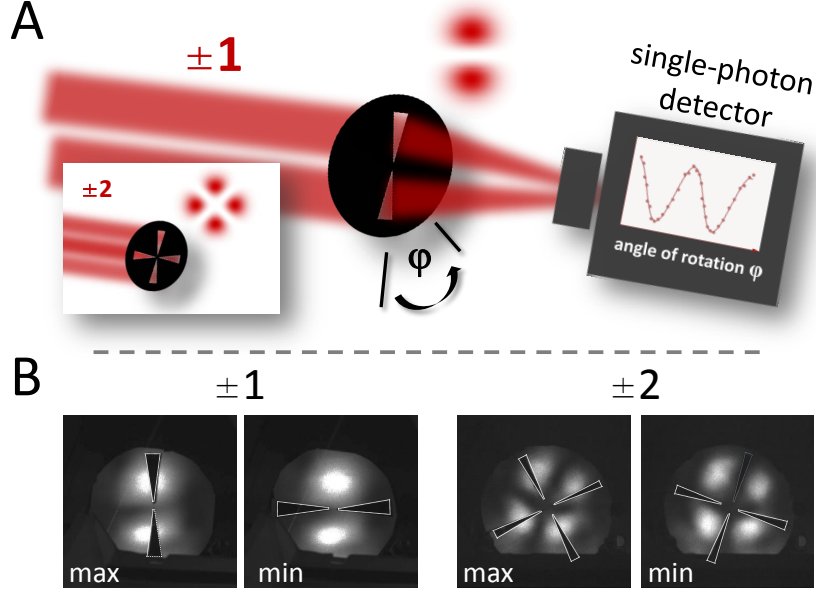


Figure 4.9: Principle of the measurement technique. The superposition of two LG modes with opposite ℓ has 2ℓ minima and maxima in a ring. The angular orientation depends on the relative phase. We use a mask, which resembles the symmetry of the beam, to measure correlations. A: An incoming beam hits the mask. For $\ell=\pm 1$, an opaque mask with two transparent slits is used to measure different superposition states. A detector after the mask collects all photons passing the slits. The superposition of $\ell=\pm 2$ has four paddles, thus we use a mask with four slits. B: Images of an alignment laser beam at the mask (mounted at the telescope at IQOQI, slits are highlighted) after 3 kilometer transmission. The laser is in a superposition of $\ell=\pm 1$ and ± 2 . The angular position of the mask is set to the maximum and to the minimum. In the entanglement experiment, we see the fringes only in coincidences.

ond TTM. To synchronize the time stamps on the two remote locations in the sub-nanosecond regime, we directly use the time-correlation of the photon pairs (which is inherently below 1ps), as explained in [139]. In addition to the usage of time-correlation, we further need to synchronize the time-tagging modules within roughly 1 second, otherwise the computation time to find a coincidence peak (as explained in figure 4.12) takes too much time. For the 1-second synchronisation, we use the *Network Time Protocol* (NTP), which is a network protocol for clock synchronization between different computers. It operates in several layers. The first layer consists of different globally distributed high-precision atomic clocks, which distribute their information to servers (so called *time servers*). The next layer consists of computers which are synchronized directly with the time servers. What follows are more layers of computers which are synchronized one layer above. The higher the layer with which a computer is synchronized, the worse is the synchronisation time accuracy. Before the experiment, we synchronize the two computers at the different locations with NTP. In all experiments, the accuracy of the synchronization was between 50 ms and 1 sec.

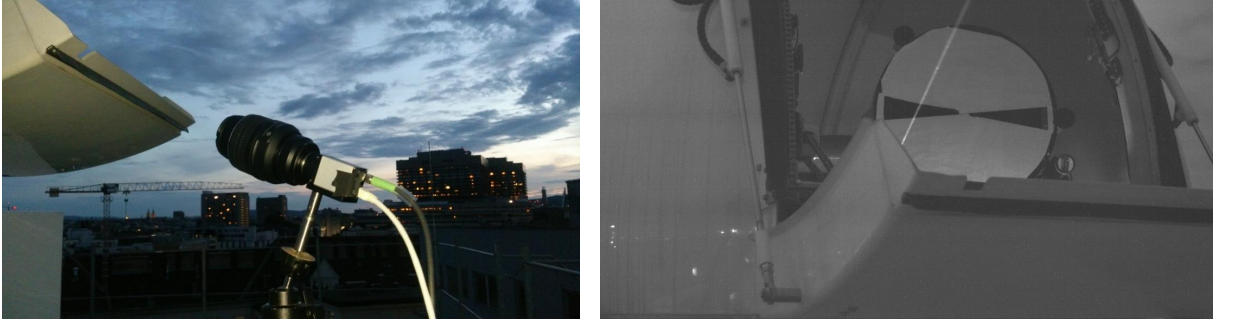


Figure 4.10: A infrared camera has been used to observe the slit-mask in front of the telescope. The image from the camera was used in real-time for alignment of the spatial modes (see figure 4.9).



Figure 4.11: A picture of the Hedy Lamarr Quantum Communication Telescope at the roof top of the IQOQI building, with the absorptive mask for $\ell = \pm 2$

4.3.2 Measurement results

In the experiment, we perform visibility measurements in two mutually unbiased bases (MUBs). For photon A, the bases are diagonal or antidiagonal and right- or left-circular polarization ($|\psi_x\rangle = \frac{1}{\sqrt{2}}(|H\rangle \pm |V\rangle)$ and $|\psi_y\rangle = \frac{1}{\sqrt{2}}(|H\rangle \pm i|V\rangle)$). For photon B, we measure in the superposition bases of two opposite OAM (specifically, $|\psi_x\rangle = \frac{1}{\sqrt{2}}(|\text{LG}_{+\ell}\rangle \pm |\text{LG}_{-\ell}\rangle)$ and $|\psi_y\rangle = \frac{1}{\sqrt{2}}(|\text{LG}_{+\ell}\rangle \pm i|\text{LG}_{-\ell}\rangle)$ – with $\ell = 1$ and $\ell = 2$ in our experiments. Here, the intensity of the superposition structure is a ring with 2ℓ intensity maxima and minima. By changing the phase of the superposition, the intensity structure is rotated. With the slit mask, we can measure photons in the σ_x and σ_y bases [44]. Specifically, we measure fringes in coincidence counts for changing angular positions of the mask. We find minima and maxima of coincidences (figure 4.13), and calculate the visibility

$$\text{vis} = \frac{\max_1 - \min_1 + \max_2 - \min_2}{\max_1 + \min_1 + \max_2 + \min_2} \quad (4.3)$$

4 Long-distance transmission of spatial modes

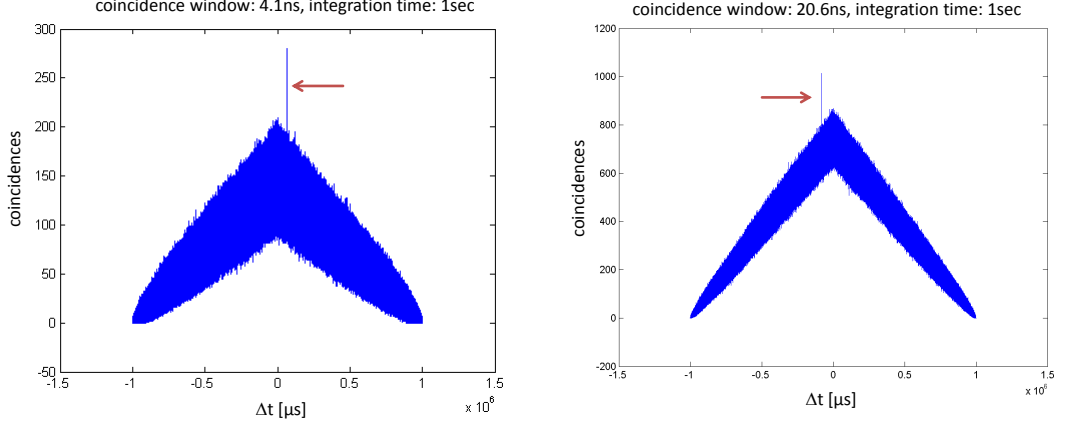


Figure 4.12: Identification of photon pairs by exploiting their time-correlation. Signals from two independent time-tagging modules are convoluted to identify a region with significantly more coincident counts. That time-information is the offset between the two TTMs. Here, convolutions from two different measurements are shown. Both are integrated over one second. In the left, coincidence windows of 4.1 nanoseconds are used, while on the right side 20.6 nanoseconds are used. The background in coincidences comes mainly from a combination of large photon countrates from the detector at the sender, and large background at the receiving detector. Small coincidence windows lead to smaller background in the coincidences (smaller number of accidental coincidence counts) but significantly longer computation time. The red arrows indicate the identified coincidence peak.

where $\min/\max_i$ are coincidence minima and maxima, which are highlighted with circles in figure 4.13. To verify entanglement, we use an entanglement witness which is the sum of the two visibilities in the two MUBs [89]

$$W = \text{vis}_x + \text{vis}_y \begin{cases} \leq 1 : \text{separable} \\ > 1 : \text{entangled} \end{cases} \quad (4.4)$$

All separable quantum states can reach at most $W = 1$, which can be understood intuitively: If a product state is perfectly correlated in one basis, it cannot be correlated in any other mutually unbiased bases. Any experimental value above $W = 1$ verifies entanglement in the system (a maximally entangled quantum state can have perfect visibility in both bases, thus $W = 2$). The visibilities are calculated directly from the maxima and minima of the measured coincidences (blue/red and yellow/green circles in figure 4.13).

In the first measurement, we use the first higher order mode with $\ell = 1$. We accumulate coincidence detections over 20 seconds at 20 different angular positions of the mask with a resolution of 9° (figure 4.13). The coincidence window is 2.5 ns. Without any corrections (such as accidental coincidence subtraction) and without any assumption about the photon statistics, we get

$$W_{\ell=1} = 1.3644 \pm 0.0084 \quad (4.5)$$

4 Long-distance transmission of spatial modes

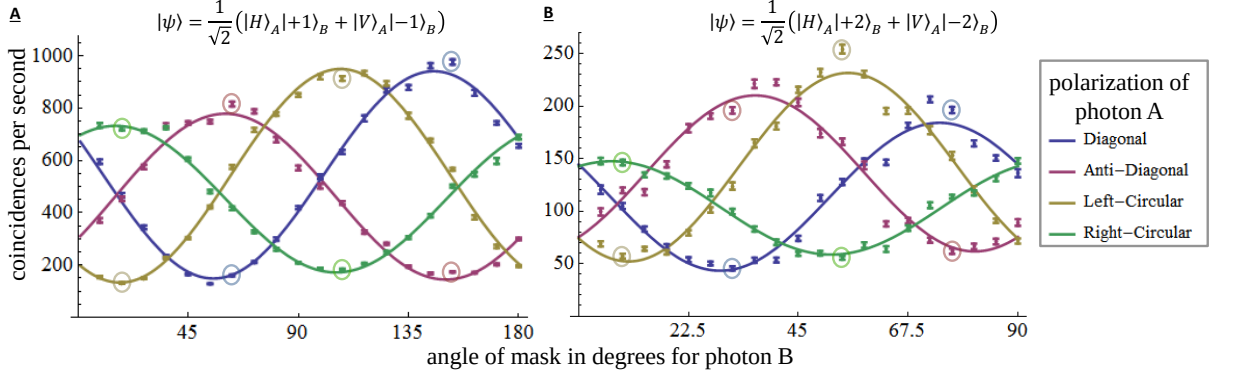


Figure 4.13: Coincidences between the transmitted photon encoded in OAM and the locally measured polarization photon. For four different polarization settings at Alice’s photon A, coincidences were recorded for 20 different angular positions of the mask at Bob’s receiver. Error bars are the standard deviation of the mean, calculated without assumptions about the underlying photon distribution from raw data by splitting the whole measurement time into 1-second intervals. The circles indicate the data used to calculate the entanglement witness. The two maxima (minima) per basis are denoted as $\max_1$ and $\max_2$ ($\min_1$ and $\min_2$) for calculating the visibility vis in the σ_x and σ_y bases. A: Raw coincidences for $\ell=1$. Coincidences for each angular position at the receiver are measured for 20 seconds. B: For $\ell=2$, we subtract accidental counts. Here, the coincidence counts for each angular position are measured for 40 seconds.

which statistically significantly confirms entanglement between the two distant photons. The error stands for the standard deviation of the mean. We calculate the error by dividing the 20 second interval at each measurement position into 20 sections of equal length, and calculate the witness eq.(4.4) for each of the 20 sections individually. From the resulting 20 values for $W_{\ell=1}$

$$\begin{aligned}
 W_{\ell=1} = & \quad 1.4354, 1.4214, 1.3942, 1.3940, 1.3928, \\
 & \quad 1.3858, 1.3848, 1.3841, 1.3804, 1.3692, \\
 & \quad 1.3633, 1.3560, 1.3522, 1.3472, 1.3366, \\
 & \quad 1.3361, 1.3346, 1.3279, 1.3173, 1.2743,
 \end{aligned} \tag{4.6}$$

we calculate the mean value and its uncertainty. In order to calculate the uncertainty of $W_{\ell=1}$, we did not need to assume any specific photon statistics. In many cases, Poissonian distribution is a good approximation of the photon statistics. However, it neglects additional sources of fluctuations, which can become relevant in experiments without controlled environments, such as free-space experiments. If we had assumed Poissonian distribution in our experiment, we would have underestimated the uncertainty significantly by around 70%, which is mainly due to atmospheric turbulence and instabilities at the sender. If we subtract accidental coincidences ($\approx 85 \pm 3$ counts/sec), the average visibility in both bases is roughly 84.2%. The visibility is enough to violate a Bell-type inequality, which violates local realism and offers the possibility of entanglement based quantum key distribution.

4 Long-distance transmission of spatial modes

In a second experiment, we transfer the photon to $\ell=2$ before transmission, send it to the receiver and measure coincidence counts for 20 different mask positions, each 4.5° rotated, for 40 seconds per setting. Here we get

$$W_{\ell=2} = 1.139 \pm 0.021 \quad (4.7)$$

verifying entanglement with $\ell=2$. Again, the error stands for the standard deviation of the mean, which has been evaluated equivalently as before: 40 seconds measurement intervals are divided into 40 parts of 1 second length. It results in 40 independent values of $W_{\ell=2}$:

$$\begin{aligned} W_{\ell=2} = & \quad 1.4218, 1.4201, 1.4106, 1.3708, 1.2581, \\ & \quad 1.2570, 1.2452, 1.2338, 1.2245, 1.2180, \\ & \quad 1.2158, 1.2142, 1.2055, 1.2051, 1.2023, \\ & \quad 1.2023, 1.1919, 1.1738, 1.1454, 1.1312, \\ & \quad 1.1235, 1.1091, 1.0995, 1.0950, 1.0778, \\ & \quad 1.0702, 1.0695, 1.0524, 1.0492, 1.0410, \\ & \quad 1.0283, 1.0248, 1.0230, 1.0190, 1.0141, \\ & \quad 0.9927, 0.9926, 0.9789, 0.8963, 0.8635, \end{aligned} \quad (4.8)$$

from which the mean and its error was obtained. Note that again we did not assume any information about the photon statistics. However, we had to subtract accidental coincidence counts ($\approx 95 \pm 7/\text{sec}$, and can be precisely obtained by the time-tags without performing any additional measurement, as shown in figure 4.14), because the signal-to-noise ratio was significantly larger (5,5% for $\ell = 2$ compared to 36,0% for $\ell = 1$), which can be understood from the different weather conditions during that night (see below for detailed weather information), different detection efficiencies at the telescope and smaller slit size of the masks (see below for detailed detection count rates).

As we collect timing information of the photons at the sending and receiving stations, we can access the number of accidental counts directly: At the offset between the time-tagging clocks, which correspond to the arrival times of the photons from a pair at the two locations, real coincidence counts from the entangled pairs are found. At every other offset, accidental coincidences can be seen (figure 4.14).

Weather conditions during the measurement nights

Figure 4.15 depicts the detailed weather data from the measurement nights. Specifically important is figure 4.15A the meteorological visibility. It is a measure of the distance at which a black object can be clearly identified from the background and is defined as the distance x_v where the contrast $C_v(x) = \frac{F_{BG}(x) - F_0(x)}{F_{BG}(x)} = 0.02$. F_{BG} and F_0 are intensities of the background and the black object, respectively. At the position of the object (at $x=0$) $F_0(0) = 0$, thus $C_v(0) = 1$. Under very good conditions, x_v can reach $\approx 100\text{km}$. In our experiment, the meteorological visibility is a measure both of loss and background light. The loss can also be caused by large values of humidity, and background light is increased for low cloud base (figure 4.15F). The change in temperature (figure 4.15B) as well as the average and maximum wind (figure 4.15D,E) are influencing the turbulences of the atmosphere.

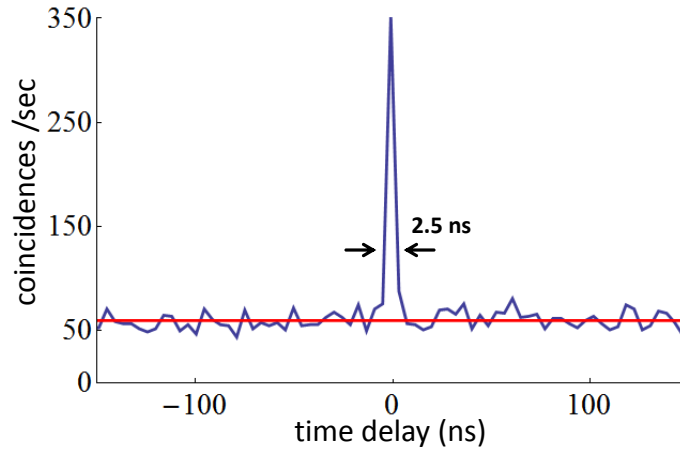


Figure 4.14: Example for measured coincidence counts from entangled photon pairs and accidental coincidence counts. The blue line indicates the coincidences calculated by overlapping the time-tagging files from the two locations. If the time delay is zero, real coincidences can be seen. For wrong time delays, only accidental counts are visible. The red line shows the average accidental counts.

In the measurement of entanglement involving $\ell=2$, the meteorological visibility was significantly smaller which was one reason for the lower fringe visibility in figure 4.13. Temperature, wind speed, humidity and cloud base were similar in the two nights.

Mode misalignment at mask

The detection method used here is axis-dependent, which means that the mode and the mask have to be aligned well (see figure 4.9B). Misalignments such as shifts of the mode relative to the mask reduce the observable visibility. The misalignment can be introduced by first-order atmospheric influences such as tip and tilt. The effect can be simulated (figure 4.16), and we find that higher modes are more sensitive to alignment, which is one reason why the observed visibility for our $\ell = \pm 2$ measurement was lower than for the $\ell = \pm 1$ measurement.

In the experiment with $\ell=1$, we had an average visibility of roughly 68,2%. With accidental subtraction we have 84,2% visibility, and accounting for non-perfect visibility from the entanglement source (which was roughly 97,5%), and imperfect polarization-compensation in fibers we would expect a visibility of $\approx 89,0\%$. The remaining decrease in visibility could be explained by atmospheric turbulence. The first order effect is a tip-tilt, which reduces the visibility as seen in figure 4.16. The loss of $\approx 11\%$ can be explained by an average misalignment of the beam relative to the mask by 25% of the beam waist. For higher-order modes the reduction is more prominent. However, this effect could be compensated with readily available adaptive optics systems in the future.

Signal and background counts

In the measurement of $\ell = 1$ and $\ell = 2$, we detect on average the following single-photon counts at the sender (which is measured in the polarization degree of freedom):

4 Long-distance transmission of spatial modes

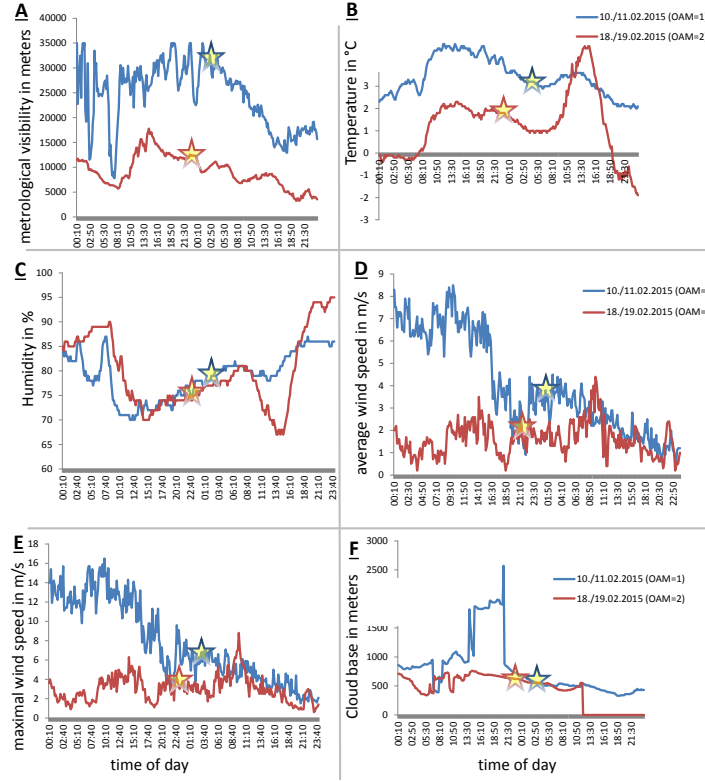


Figure 4.15: Detailed weather conditions of the two measurement nights. Blue (red) curve stands for the nights we measured entanglement involving $\ell=1$ ($\ell=2$). The blue (red) star indicated the precise time. A shows the meteorological visibility, B shows the temperature, C shows the humidity, D shows the average wind speed, E shows the maximal wind speed and F shows the cloud base.

Sender (counts per Arm)			
ℓ	Singles/sec	Background/sec	Signal/Noise
$\ell = \pm 1$	800.000	10.000	79
$\ell = \pm 2$	770.000	10.000	76

At the receiver, we find the following count rates:

Receiver			
ℓ	Singles/sec	Background/sec	Signal/Noise
$\ell = \pm 1$	36.600	27.800	0,36
$\ell = \pm 2$	40.100	38.000	0,055

In the measurement of entanglement with $\ell = 2$, we needed to subtract accidental coincidence counts. The reason was a large number of background counts compared to the small number of photons from the entangled pairs, with a signal-to-noise ratio of 0,055. The smaller ratio (compared to $\ell = 1$) can be understood by the smaller meteorological visibility (as explained above), less efficient collection at the telescope (figure 4.18) and a smaller slit opening angle of the mask.

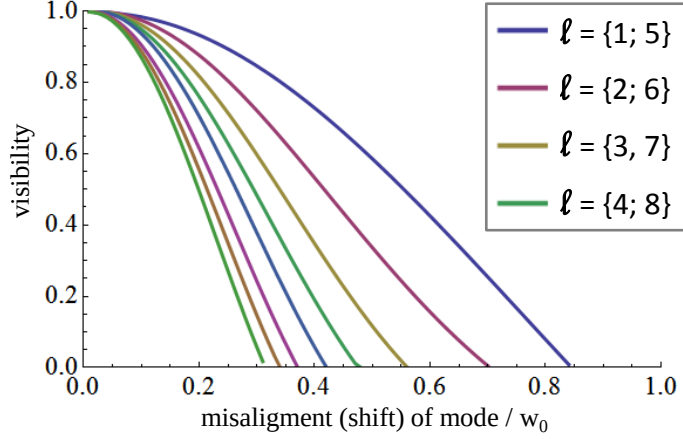


Figure 4.16: Visibilities of superpositions of $\ell = \pm 1$ to $\ell = \pm 8$ are plotted as a function of the relative misalignment of the mode and the mask, in units of the beam waist w_0 . $\ell = \{1; 5\}$ means that blue stands for $\ell=1$ and $\ell=5$, the most left curve is $\ell = 1$. The higher-order mode is more sensitive to misalignment, which means the visibility drops faster. The visibilities are obtained by simulating a misaligned mode at a mask, and fitting the result with the expected sinusoidal function. The experimentally obtained visibility can be used to upper bound the potential misalignment of the mask in the experiment. For $\ell = 1$, we maximally misaligned the modes by $0.55 w_0$, while for $\ell = 2$ the maximal misalignment was $0.45 w_0$ – assuming that everything else was perfect.

Estimation of accessible OAM channels

As a final measurement, we estimate the number of different orthogonal quantum channels we have access to in principle. As OAM modes grow for higher numbers of ℓ and our receiver telescope has a finite size, there is a maximum number of ℓ that can be detected. For that, we transfer photon B to different values of ℓ (from $\ell=0$ to 15) and record the number of coincidences with photon A. Thus, photon A is a trigger for the higher-order ℓ -modes of photon B after sending it across the city. More precisely, we create the state

$$|\psi\rangle = \frac{1}{\sqrt{2}} (|H, 0\rangle_A |V, 0\rangle_B + |V, 0\rangle_A |H, +\ell\rangle_B). \quad (4.9)$$

where A stands for photon A, which is detected at the sender, and B stands for photon B which is sent across the city to the receiving telescope. The first term $|H, 0\rangle_A |V, 0\rangle_B$ is used to actively synchronize the time-tagging modules at the two locations, again with the method presented in [139]. The measurements for the second term are shown in figure 4.17: Whenever the photon at the sender is vertically polarized, we know that we transmitted photon B with ℓ , where ℓ goes from 0 to +15.

Here, no mask is in front of the telescope. The detected coincidence rates in figure 4.17 show that photons up to $\ell=5$ can be distinguished from the background. The graph can very well be described by the geometry of our telescope, which cuts the incoming beam both at the primary and secondary mirror. Counts of high order ($\ell \leq 10$) can't be distinguished from the background. With our sender and receiver, we have access to roughly 11 quantum channels of

4 Long-distance transmission of spatial modes

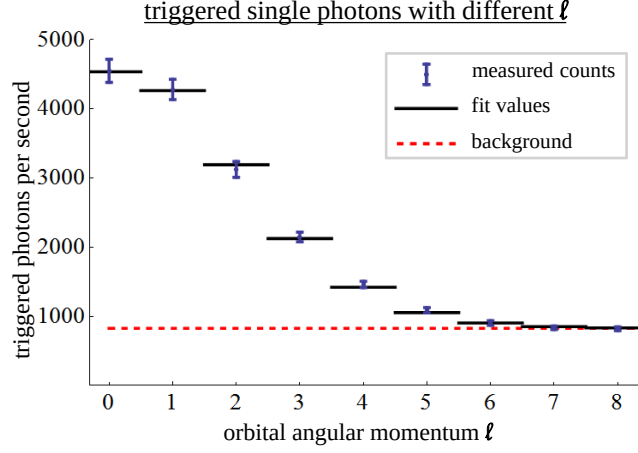


Figure 4.17: Triggered single-photon counts for different orbital angular momentum ℓ . We use correlated photon pairs (blue, each point is measured for 60 seconds) to determine the number of accessible OAM modes at the telescope. For that, we measure one photon at the sender, and transfer the correlated partner photon to a higher-order OAM mode (from $\ell=0$ to $\ell=8$), which is then transmitted to the telescope 3 kilometers away. The lower rate of coincidence counts for higher-order modes is due to the geometric restrictions (finite size of primary and secondary mirror) of the telescope, which can be modeled very well (black line). The error bars show the standard deviation. The red dashed line indicates the background counts (calculated from average counts of $\ell=10$ to $\ell=15$). The data shows that we are able to access modes up to $\ell=5$ from the background, which constitutes 11 orthogonal quantum channels ($\ell=-5$ to $\ell=5$).

OAM ($\ell=0$ to $\ell = \pm 5$).

The background in figure 4.17 originates mainly from two effects: The first effect is accidental coincidence counts because of large count rates (1.500.000 single counts/sec at the receiver, which lead to 525 ± 10 accidental counts/sec). The second effect is due to misidentification of polarization in the transfer-setup (170 ± 25 counts/sec). If the polarization changes between the source and the transfer-setup (which is likely because fibers were used, which are not compensated perfectly), some photons are not modulated by the Spatial Light Modulator and thus end up in a Gaussian state, which are detected by the telescope.

The expected count rates depend on a combination of the received modes and the geometry of the receiver telescope. The telescope clips the beam in two different ways, which results in a reduced number of detected photons: Firstly, the primary mirror has a finite diameter of roughly 37 cm. Secondly, the secondary mirror of the telescope is in the centre of the beam path. The Gauss mode has the maximal intensity there, thus it is substantially cut at the secondary mirror. The effect can be illustrated graphically in figure 4.18.

In the experiment, we prepare OAM-modes with phase-only holograms. The size of those modes scale linear with ℓ . There is no closed-form solution for such holograms, thus we calculate them numerically by applying a Fourier transformation of a Gaussian beam with a helical phase (which is introduced by the SLM):

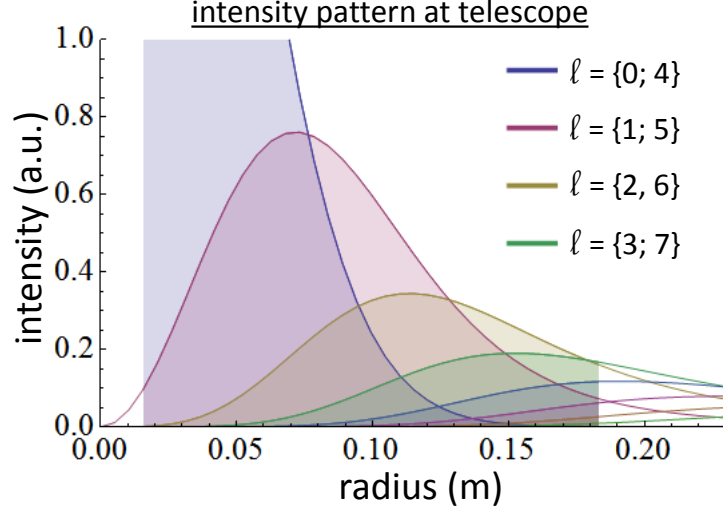


Figure 4.18: The blue, red, yellow and green curves present the radial intensity distribution of OAM modes $M_\ell(r, \phi = 0)^2$ ($\ell=\{0;4\}$ means that blue stands for two different modes, $\ell=0$ and $\ell=4$). The filled region indicates the part of the beam that arrives at the photon detector. It is limited on the inside by the shading of the secondary mirror and on the outside by the finite size of the primary mirror.

$$M_\ell(r, \phi) = N \cdot \mathcal{F}\left[e^{-\frac{r^2}{w_0^2}} \cdot e^{i\ell\phi}\right] \quad (4.10)$$

Here, $\mathcal{F}[\cdot]$ stands for the Fourier-transform, w_0 is the gaussian beam-waist and N is a normalization constant. The expected intensities for different modes can be easily calculated with the given boundary conditions. The function

$$F(\ell) = \text{bg} + \text{cnt} \cdot \int_0^{2\pi} d\phi \int_{SM}^{PM} r \cdot M_\ell(r, \phi)^2 dr \quad (4.11)$$

is used to obtain the fit in figure 4.17, with cnt (counts), SM (size of secondary mirror), and w_0 (beam waist at telescope) being the fit-paramters. Here, the background bg has been calculated from the measured data (the mean of the counts from $\ell=10$ to $\ell=15$), and PM (radius of primary mirror) has been measured to be roughly 18,5 cm. The obtained fit value resemble very well the physical dimensions of the telescope (radius of the secondary mirror is SM=1,2cm) and of the observed classical alignment beam ($w_0=8,5\text{cm}$) in figure 4.9.

4.3.3 Remarks

Here we have shown that even individual photons can retain the information of the spatial structure after transmission over several kilometers across a city. It shows that both the spatial coherence of the individual photons has been preserved, and that the coherence of two-photons after the transmission can still be observed.

With these results it is shown that quantum experiments with spatial structures of light are quite possible - contrary to previous theoretical and numerical predictions.

A logical next step is to exploit the potential of high-dimensional entanglement, as shown in chapter 3. The question arises whether and how high-dimensional entanglement can be demonstrated with a similar methods as demonstrated here. The great advantage of our method is that one does not have to make a holographic transformation at the receiver, which is very susceptible to influence of turbulence. One could use single-photon sensitive cameras (such as ICCD cameras [138]), and investigate similar encoding as shown in [140] (there, multiplexing of many spatial modes has been created and detected with classical cameras and image recognition algorithms). For that, well controlled high-dimensional quantum states are required, in particular maximally d -dimensional entangled states and no contributions from $(d+1)$ -dimensional modes. This could be done by shaping the pump beam, as shown in [141]. An alternative method has been developed has been developed in chapter 5.6, which is based on the identification of the paths of photons which removes the *which-crystal*-information without erasing it explicitly.

Another method to transmit and recover high-dimensional entanglement is the exploitation of adaptive optics systems, which have already been thoroughly investigated for classical communication with OAM [142]. One significant contribution of the atmosphere are shifts of the beam axis (which is the first order contribution in the Zernike basis, as explained above). Such beam shifts could be corrected with fast tip-tilt mirrors, in order to allow holographic transformations.

4.4 Classical transmission over 143 kilometers

The content of this chapter is based on the publication "Twisted Light Transmission over 143 kilometers" [111]⁴.

The second follow-up experiment from the work shown in chapter 4.2, we investigate potential limits of the distance, at which communication with spatially encoded light beams is still possible. This question is relevant to understand the potential of spatial modes in communication (both classical and quantum) in real-world environment. We have shown in 4.3 that within a 3-kilometers link across Vienna, single photons robustly carry the encoded spatial mode information which can be measured at the receiver. Are larger distances realistic? For instance, can one communicate across larger cities with distances of 10 kilometers? Can one communicate between two cities that are dozens of kilometers apart? Could one communicate with satellites (not taking into account the beam diameter of such modes for the moment)? Could one even transmit and receive spatially encoded modes over more than 100 kilometers, for instance between two islands?

In this chapter, we investigate the last of these questions, and analyse the properties of modes propagating over 143 kilometers between two Canary Islands, La Palma and Tenerife. By that, we increases the distance from 3 kilometers to 143 kilometers, which is almost a factor of 50. As before, we use OAM mode superpositions of $\ell = \pm 1, \pm 2$ and ± 3 with different relative phases for encoding information. The different phase results in a different rotation of the mode, which makes it possible to distinguish these light beams by their intensity. For characterization of the

⁴Mario Krenn, Johannes Handsteiner, Matthias Fink, Robert Fickler, Rupert Ursin, Mehul Malik, Anton Zeilinger, Twisted Light Transmission over 143 kilometers. *PNAS* **113**, 13648–13653 (2016).

received mode quality, we record images of the intensity distribution observed on the white wall of the telescope *Observatorio del Teide* and analyze them with a pattern recognition algorithm based on an artificial neural network, which is explained in chapter 4.2.2. By calculating the cross-talk, we find that the modal structure can be distinguished quite well even without the use of any adaptive optics correcting for the effects of atmospheric turbulence. Finally, to visualize the quality of the transmission, we use these modes to encode, transmit, and recover a short message.

4.4.1 Experimental setup

In the experiment, we use the superpositions of OAM modes. Equally weighted superpositions of LG modes with opposite OAM can be written (in the same way as in eq.(4.1)) as

$$|\text{LG}_{\pm\ell}^{\alpha}\rangle = \frac{1}{\sqrt{2}} (|\text{LG}_{+\ell}\rangle + e^{i\alpha}|\text{LG}_{-\ell}\rangle), \quad (4.12)$$

where α denotes the relative phase between the two modes. The transverse phase is radially uniform and has 2ℓ phase jumps of π in the azimuthal direction, which leads to 2ℓ maxima and minima arranged symmetrically in a ring. The phase α is directly related to the angular position of the structure $\gamma = \frac{360^\circ}{2\pi} \frac{\alpha}{2\ell}$. The angular position can reveal the relative phase in the superposition, a feature that has been explained in chapter 4.2 in the classical context, and in chapter 4.3 in the quantum context.

A schematic of the experimental setup for sending and receiving the spatial modes can be seen in figure 4.19a. At the sending station on La Palma, a green laser with a wavelength of 532nm and a power of 60mW was used for encoding the twisted light modes and their superpositions. The laser was modulated with a phase-only spatial light modulator (SLM), which imprinted the spatial modes on the beam without amplitude modulation. Phase-only holograms, as used by us, produce vortex beams with a slightly different radial structure compared to LG modes. This difference is not relevant in our experiment. Then, the beam was magnified with a telescope to approximately 4 cm diameter, and transmitted with a high-quality $f = 30$ cm lens through 143 km of free space to the island of Tenerife, where the receiver was located. In vacuum, the beam diameter at the receiver would be expected to be roughly 1.3 meters for a Gauss mode, while we observe significantly larger beams due to beam spreading introduced by the atmosphere. At the receiver in Tenerife, the mode structure (with a diameter of roughly 11 meters) was observed on the white wall of an observatory and recorded with a NIKON D3S camera with varying exposure times. A beacon laser shining back from Tenerife to La Palma was used for initial alignment. However, no active tracking or adaptive optics was used for correcting the atmospheric turbulence during the data transmission itself. The recorded images were then analyzed to recover the encoded information in an automatized manner.

Figure 4.19b shows the sending setup in very strong turbulence conditions on the island of La Palma. Vortices and eddies formed by water vapor droplets in the air can be clearly seen in the inset. We were unable to recognize any spatial modes transmitted during such atmospheric conditions. The long-time exposure photograph in Figure 4.19c was taken under much better atmospheric conditions and shows an $\ell = \pm 1$ superposition being transmitted from La Palma to Tenerife, over the edge of the crater wall of the Caldera de Taburiente. The insets show that the double-lobed structure of the mode is clearly visible in the transmitted beam.

4 Long-distance transmission of spatial modes

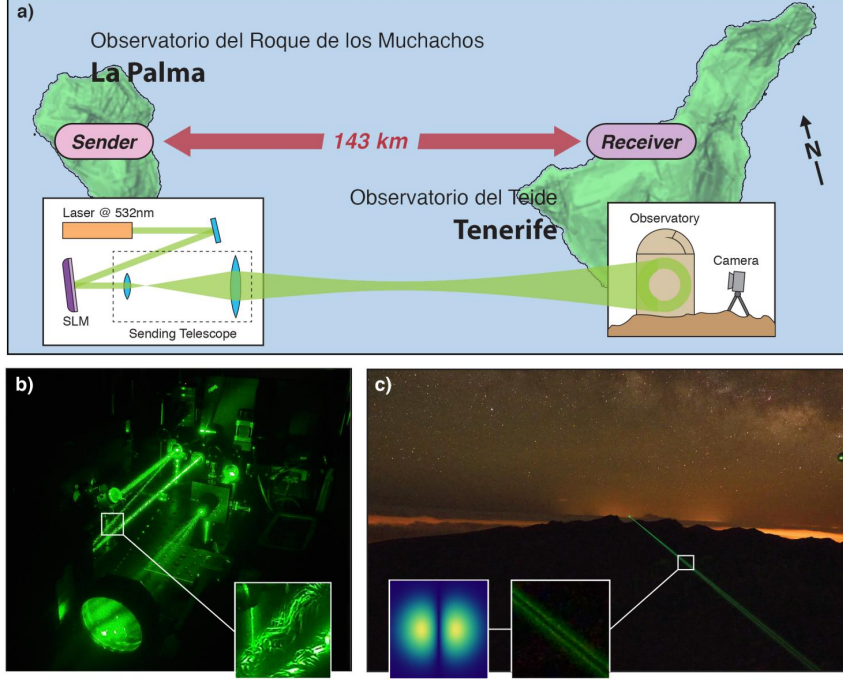


Figure 4.19: **a)** Sketch of the experimental setup. The sender is located on the roof of the *Jacobus Kapteyn* telescope on the island of La Palma and consists of a 60mW laser with a wavelength of 532nm modulated by a spatial light modulator (SLM). Different phase holograms on the SLM encode different spatial modes. The modes are magnified with a sending telescope and sent over 143 km to the receiver at the island of Tenerife. Extra mirrors used in the actual sending setup are not shown for simplicity. The structure of received modes is observed on the wall of the telescope *Optical Ground Station* (OGS) owned by the European Space Agency and recorded with a camera. **b)** A photo of the sender taken during extremely turbulent conditions on La Palma. Small vortices and eddies formed by the water vapor in the air are clearly visible in the inset. At the sending lens an $\ell = \pm 1$ can be seen. Modes sent under these conditions were not discernable at the receiver. **c)** Long-time exposure photo showing an OAM superposition of $\ell = \pm 1$ being transmitted over the Caldera de Taburiente (silhouetted in black) from La Palma to Tenerife. The insets show that the double-lobed modal structure of the beam is clearly visible. A theoretical plot of the mode superposition cross-section is shown for comparison.

To characterize the turbulence, we took long-time exposure photographs of a Gaussian beam shining back from the receiver towards the sender at La Palma and investigated the observed beam spread. Without atmosphere one would expect a Point Spread Function (PSF) with a Full Width Half Maximum (FWHM) of $FWHM = \frac{\lambda}{D}$, where D is the effective aperture of the objective. Propagation through turbulent media enlarges the diameter of the PSF, which leads to a $FWHM_{obs}$, that can be characterized by the Fried parameter r_0 that is defined by $r_0 = \frac{\lambda}{FWHM_{obs}}$. We used an Olympus E-M10 camera and an objective with an f-stop of f/5.6 and focal length $f = 200\text{mm}$ to record images of the tracking beam at La Palma. The effective aperture of this system was $D=35\text{mm}$ and therefore twice as big as the expected r_0 . During the

observation period, the Fried parameter varied between 0.4 cm and 1.3 cm, which is consistent with measurements over the same link from earlier years [125] and is considered to demonstrate strong turbulence.

4.4.2 Transmission quality

We performed the measurements over ten successive nights. On four of these nights, we analyzed data for mode superpositions, and on two nights, we recorded individual vortex modes (not superpositions). On the remaining four nights, the weather conditions were too bad in order to recognize any mode structure. This was either because of fog or clouds between the two islands that significantly reduced the received laser intensity, or because of the presence of strong turbulence that significantly deteriorated the mode quality. In these cases, the Fried parameter r_0 of the link was below 1 cm.

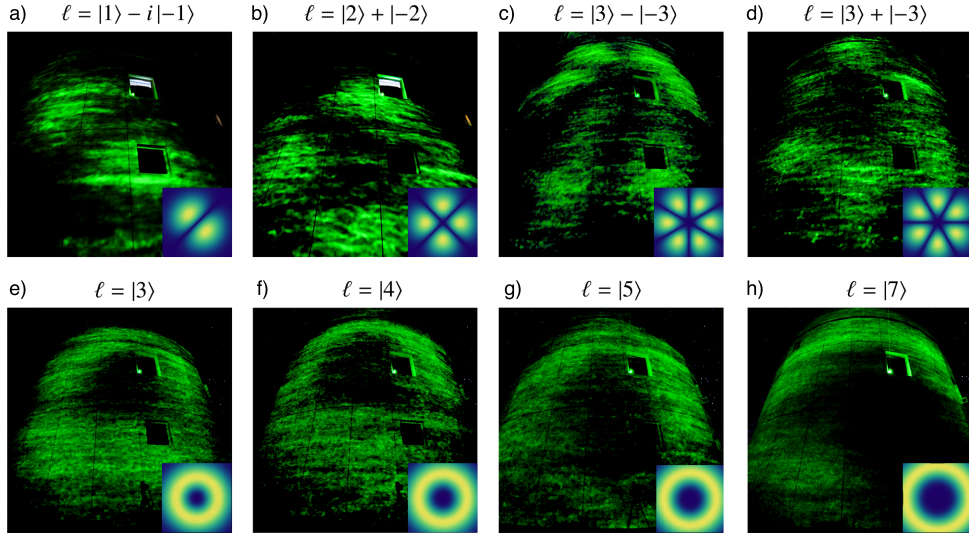


Figure 4.20: a)-d) Examples of OAM mode superpositions received on the wall of the telescope *Observatorio del Teide* after propagating through 143 km of free space between the islands of La Palma and Tenerife. The diameter of the observatory is roughly 11 meters. The lobed modal structure is clearly visible for mode superpositions with $\ell = \pm 1, \pm 2$, and ± 3 . Images c) and d) show the rotation of a $\ell = \pm 3$ mode superposition by $\frac{\pi}{3}$ when the relative phase α is changed by π . e)-h) Examples of pure OAM (vortex) modes observed at the receiver. The intensity null at the center of the modes is clearly visible. The mode diameter gets larger as the OAM quantum number ℓ is increased, and is seen to approach the size of the telescope wall for $\ell = 7$. The size of the modes clearly increases for higher orders. Note that these images were taken at a time when atmospheric conditions were stable. Furthermore, the mode information can be extracted much more reliable from the superposition modes (upper row) than from the OAM modes themselves (lower row).

When the Fried parameter r_0 was above 1 cm, we were able to distinguish the OAM modes received at Tenerife. Examples of four of such OAM mode-superpositions as well as four vortex modes are shown in figure 4.20. The lobed modal structure is clearly visible for modes with

4 Long-distance transmission of spatial modes

	0°	22.5°	45°	67.5°	90°	112.5°	135°	157.5°
0°	24	1	0	0	0	0	0	0
22.5°	3	22	0	0	0	0	0	0
45°	0	0	19	2	2	0	0	4
67.5°	0	0	0	26	4	0	0	0
90°	0	0	0	7	21	1	0	0
112.5°	0	0	0	0	2	21	0	2
135°	0	0	0	0	1	4	11	8
157.5°	1	0	0	0	0	0	0.5	23.5

Table 4.1: Cross-talk matrix from figure 4.21A for $\ell=\pm 1$ modes with different rotations. Sent and detected modes are written vertically and horizontally, respectively. Modes with different relative orientations are sent, which correspond to a relative phase between the $\ell=+1$ and $\ell=-1$ term. For example, for 25 images of orientation 0°, the neural network identified 24 of them correctly, whereas one image has been identified as orientation of 22.5°. For sent modes with orientation of 157.5°, one image was identified with same likelihood of 135° and 157.5°, thus the 0.5 and 23.5.

$\ell = \pm 1, \pm 2$, and ± 3 in figure 4.20a-d. The relative phase of π introduced in the $\ell = \pm 3$ mode superposition is clearly seen to rotate the mode structure by an angle of $\frac{\pi}{3}$ in figure 4.20c and 4.20d. Examples of four different pure vortex modes ($\ell = \pm 3, 4, 5$, and 7) received after 143 km are shown in Figs. 4.20e-f. As expected, the mode diameter gets larger as the OAM quantum number ℓ is increased, and is seen to approach the size of the telescope wall for $\ell = 7$.

In the next step, we estimate how the transmission affects the relative phase in the superposition. The advantage of our method is that the relative phase of LG superpositions is visible directly from intensity measurements for a fixed value of ℓ , thus allows us to estimate their quality in the transmission from the recoded images alone. The mapping of observed intensities to the relative-phase information was done with a pattern recognition algorithm which, for every image, returns the most likely relative phase: The algorithm is based on an artificial unsupervised neural network, also known as a self-organizing feature map [123]. The idea has been used by us in [109] (chapter 4.2) and is explained in detail in the earlier chapter. Basically, one supplies the neural network with a training set of images. The images are then characterized automatically according to their respective features. After the training phase, the network can analyze real data in the form of images. The training set consists of images that were sent through the same turbulent link, allowing the algorithm to automatically find a robust characterization of images of modes exposed to turbulence.

Using this detection method, we are able to calculate cross-talk matrices between the sent and received modes. The cross-talk matrix shows to what quality the neural network was able to correctly identify the received image. (More details in the appendix.) We analyze superposition structures and their angular rotations, which correspond to different relative phases $\Delta\alpha$ between the modes (see figure 4.21, upper row and Table 4.1). For $\ell = \pm 1$ and eight different settings of $\Delta\alpha = \frac{\pi}{4}$, nearly 80% of the different rotation angles of the structure were correctly identified. For $\ell = \pm 2$ with four different settings of $\Delta\alpha = \frac{\pi}{2}$, 84% of the mode images were correctly identified. For superpositions made up of $\ell = \pm 3$, two different relative phases $\Delta\alpha = \frac{\pi}{2}$ were

4 Long-distance transmission of spatial modes

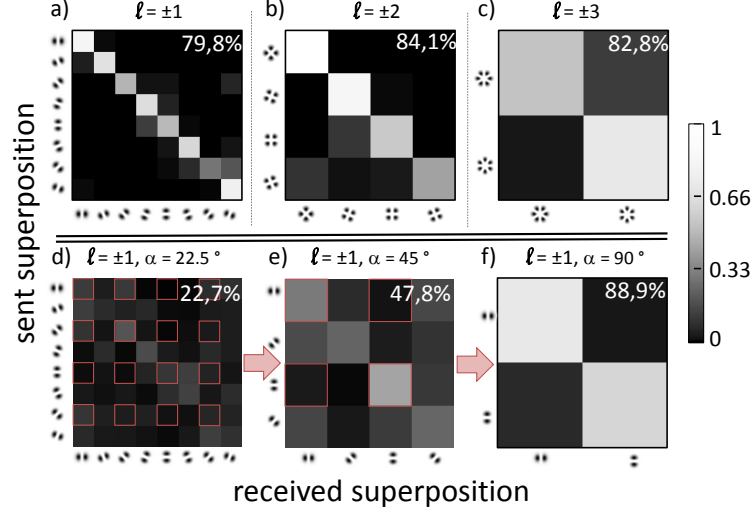


Figure 4.21: Cross talk matrices showing the success probability with which the transmitted OAM mode superpositions were correctly identified at the receiver. **a)-c)** Results for superpositions of $\ell = \pm 1$, ± 2 and ± 3 modes with relative phases of $\Delta\alpha = \frac{\pi}{4}, \frac{\pi}{2}$ and π , respectively (with 8, 4 and 2 settings of the phase, respectively). The different relative phases correspond to different rotations of the superposition structure. The received modes were correctly identified by our detection algorithm with an average success probability of 82%. **d)** During a turbulent night, 8 modes consisting of superpositions of $\ell = \pm 1$ were identified with a success probability of only 22.7%. When we restrict ourselves to a subset of these modes with $\Delta\alpha = \frac{\pi}{2}$ or π (highlighted with red squares and reanalyzed by the neural network each time – which means, restricting the alphabet in the communication), the success probability increases significantly. The ability to resolve all eight of these modes is required for quantum key distribution based on violation of Bell’s inequality, **e)** four modes are necessary for entanglement-based quantum key distribution (violation of an entanglement witness), **f)** two modes can be used for classical communication with one bit per mode.

identified with nearly 83% certainty. Details to all measurements can be seen in Table 4.2.

From this, a channel capacity can be calculated of 2,24 bits, 1,45 bits and 0,39 bits per shot, for $\ell = \pm 1$, ± 2 and ± 3 respectively. On certain nights, the atmospheric conditions did not allow small phase changes to be identified very well, as is seen by the success rate of 22.7% for $\ell = \pm 1$ and $\Delta\alpha = \frac{\pi}{4}$ (figure 4.21d). By restricting ourselves to a smaller subset of these modes, we were able to increase the success rate to 47.8% and 88.9% for $\Delta\alpha = \frac{\pi}{2}$ and $\Delta\alpha = \pi$, respectively. Verifying the presence of OAM entanglement requires one to measure different mode superpositions with at least $\Delta\alpha = \frac{\pi}{2}$. This indicates that in certain conditions where quantum communication would break down (as it needs four different phase settings), classical communication using OAM states might still be possible (with a two-level alphabet). By analyzing intensity images, we estimate the expected visibility in a quantum entanglement experiment (in a setup similar to that explained in chapter 4.3) to be 60%, which indicates that the distribution of quantum entanglement over this link encoded in spatial modes is not

4 Long-distance transmission of spatial modes

	$\ell=\pm 1$ 12.04., 01:45	$\ell=\pm 1$ 12.04., 02:45	$\ell=\pm 1$ 15.04., 23:45	$\ell=\pm 2$ 16.04., 00:15	$\ell=\pm 1$ 19.04., 01:45	$\ell=\pm 3$ 21.04., 02:30
Δ angle	45°	22.5°	22.5°	22.5°	45°	60°
Exposure time	0.5s	0.5s	0.5s	1s	0.25s	0.33s
#(images)	198	1024	250	133	240	115
#(training)	20	120	40	20	20	16
Duration	5 min	23 min	21 min	8 min	9 min	3 min
r_0 of zenith	9.3cm	10.2cm	15.1cm	13.8cm	11.6cm	11.1cm
r_0 of link	–	–	1.1cm	1.1cm	1.0cm	1.2cm
recognized %	58.1%	22.7%	79.8%	84.1%	74.1%	82.8%

Table 4.2: Details for measurements of cross-talk matrices. The relative phases correspond to different rotations of the superposition structure. " Δ angle" stands for the physical rotations between the different modes and corresponds to the phase in the superposition. "Exposure time" is the time we record the screen with the mode structure for the data set. "#(images)" is the number of total images of the data set. "#(training imgs)" is the number of images that were used for training the neural network. "Duration of series" is the time between the first and last image of the data set. " r_0 of zenith" is the Fried parameter r_0 in the direction of the zenith, measured at Tenerife by Robotic Differential Image Motion Monitor. r_0 of link is the horizontal Fried parameter between the two islands, over the 143-km link, recorded by using a 523-nm laser. "Recognized %" stands for the percentage of correctly identified modes by the neural network in the particular data set.

prevented by atmospheric turbulence.

4.4.3 Data transmission

As a final test of the transmission quality, we encoded a short message ('*Hello World!*') in modes $\ell = \pm 1$ with four different relative phases (see figure 4.22). The message was encoded using a 64-letter alphabet with upper- and lower-case letters, as well as numbers, space and exclamation mark. In this alphabet, every letter needs 6 bits of information for encoding. We encoded each letter by using three consecutively transmitted superposition modes, with each mode in one out of four phase settings. Thus, each mode carries 2 bits of information, which leads to $4^3 = 2^6 = 64$ settings.

We recorded five 1-second long exposures of every mode, resulting in a total of 180 images. Out of these, 72 images were chosen at random and used to train the neural network. The network then analyzed the remaining 108 images in order to decode the message. Each letter was redundantly encoded three times as a simple form of error-correction. The network decoded the message as "*Hello WorldP*", where the last letter contains one (out of three) wrongly detected mode. The error per letter is 8.33% (1 out of 12 letters are wrong), the error per bit is 1.4% (1 out of 72 bits are wrong). The individual modes were identified correctly with a probability of 76.3%. The complete transmission (including delimiters) took 271 seconds, which corresponds to a speed comparable to that of smoke signaling – the first form of long-distance communication in ancient times [143] or to that of communication with neutrinos [144].

4 Long-distance transmission of spatial modes

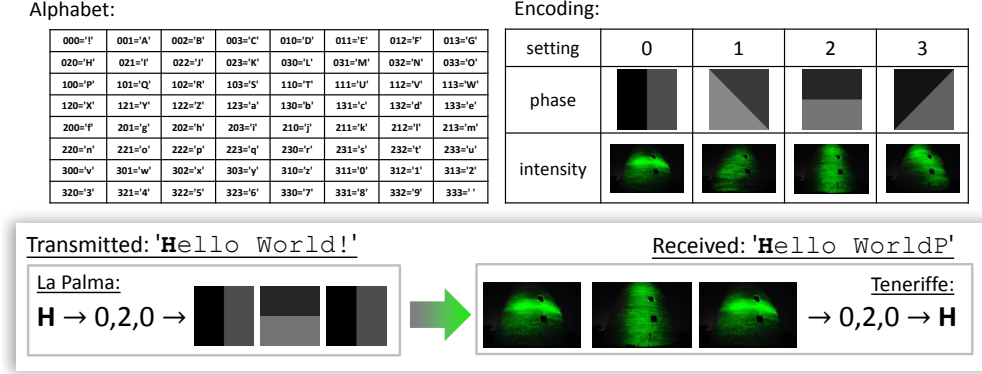


Figure 4.22: Encoding and decoding of a short message with twisted light superpositions. The message *Hello World!* is sent letter by letter. Every letter is encoded into three $\ell = \pm 1$ superpositions with four different relative phase settings. For example, the letter 'H' is encoded as 0, 2, and 0. Thus every mode corresponds to two bits of information. After 143 kilometers of transmission, the modes are recorded and characterized with an artificial neural network. The same alphabet is used to decode the letter from the mode superpositions. The final recorded message is *Hello WorldP*. The last letter is a "P" (which is encoded as 1,0,0) instead of a "!" (which is encoded as 0,0,0). This error is due to one incorrectly detected mode.

4.5 Discussion

The three experiments show that spatial modes of lights can actually be transmitted through turbulent atmosphere, and information – either classical or quantum – can be extracted after the transmission. There are several open questions which need to be investigated further. *High-speed classical communication* – Classical communication uses various degrees of freedom of light in combination to increase the number of distinguishable (orthogonal) channels. That approach, called multiplexing has been first shown with OAM modes in the laboratory in 2012 [145]. The authors use a combination of polarisation, amplitude/phase (quadrature amplitude modulation), time and OAM modes to transmit more than 1 Terabit/sec. Two years later, using also wavelength multiplexing, information transmission of 100 Terabit/sec was achieved [146]. While it has been seen that for a distance of 120 meters, corrections for the atmospheric influence are not required yet [115], one might expected that for larger distances it is necessary. Conveniently, the application of state-of-the-art adaptive optics such as those used in simple and efficient intensity-based methods [142] could further improve the link quality, and enable high-speed information transmission over large distances. An alternative approach would be to investigate the full potential of image-recognition algorithm for distinguishing modes. The advantage over conventional methods is the ability to adapt to atmospheric turbulence. In a recent study, our method of distinguishing different modes by an unsupervised neural network was expanded by more powerful neural networks (a convolutional neural network). This algorithm was used in a laboratory experiment, and showed great resistance against turbulence as well as the ability to recognize the mode information with very small intensities of light [140]. In that research, the authors have also shown the potential of distinguishing superpositions of more

4 Long-distance transmission of spatial modes

than two modes, which could be very interesting for multiplexing techniques. Investigating the potential and limits of both image recognition techniques and adaptive optics systems will be a very interesting and important to understand how encoding of information in the spatial modes of light can be used in real-world scenarios. Additionally, the size of the mode increases with larger values of the OAM, therefore larger sending and receiving telescopes will be needed. Also, more efficient encoding of the OAM modes (than we used) are known [30], which could decrease the beam size significantly. The question how much of the mode one needs to detect in order to extract the information seems to be relevant in realistic scenarios, and needs to be investigated further.

High-dimensional entanglement – We have shown that quantum entanglement can be distributed with spatial modes of photons (which also shows that both the single-photon spatial coherence as well as the two-photon coherence is not destroyed in the atmosphere). For that, we used a technique which does not require holographic transformations at a SLM, but rather uses masks that identify the spatial intensity distribution. A convenient property of LG modes allows to measure relative phases in the superposition with masks, which is sufficient to show that the state is not separable, i.e. it is entangled. It is an open question whether a similar detection scheme could reveal genuine high-dimensional entanglement. In our experiment, we started with 2-dimensional polarisation entangled and mapped it to OAM. For that reason, the measurements with masks corresponded to well-defined visibility measurements. If the photons would be high-dimensionally entangled (for instance, if their OAM-correlation was created in a SPDC process), it is more difficult to find such well-defined correlation measurements. One solution could be an entanglement source with a well-defined OAM correlation (i.e. a number of d modes are maximally entangled, and no other modes exist in the state). Such a source has been designed (or – rather, *found*) by us recently and is described in section 5.6.

In addition, another open question is whether such a technique could be used in quantum communication protocols. The only method known so far would require mode transformations, either with holograms [3] or with mode sorters [116, 117]. Both methods are very sensitive to lateral alignment. An important question is whether simple and fast tip-tilt corrections at the receiver telescope are powerful enough to correct for strong turbulence. Alternatively, adaptive measurement algorithms could improve the entanglement detection, by adjusting the measurements according to the turbulence [147].

5 Automated Search for new Quantum Experiments

The content of this chapter is based on the publication "Automated Search for new Quantum Experiments" [148]^{1 2}.

5.1 Motivation

In this final chapter, we come back to the the question of experimentally investigating complex types of quantum entanglement. The size of the Hilbert space can be increased in two ways: First, the number of dimensions in the entanglement can be increased (which has been discussed in earlier chapters). Entanglement between more than two particles has been possible only with the polarisation degree-of-freedom until recently, which is per se confined in a two-dimensional state space. Second, the number of entangled particles can be increased. In chapter 3 we have seen that photon pair can be very high-dimensionally entangled in their OAM.

Being able to create entangled states which are both high-dimensional (dimension $d > 2$) and multipartite (number of particles $n > 2$) entangled opens the possibility to explore new types (e.g. [149–151]) and new properties (e.g. [152, 153]) of entanglement.

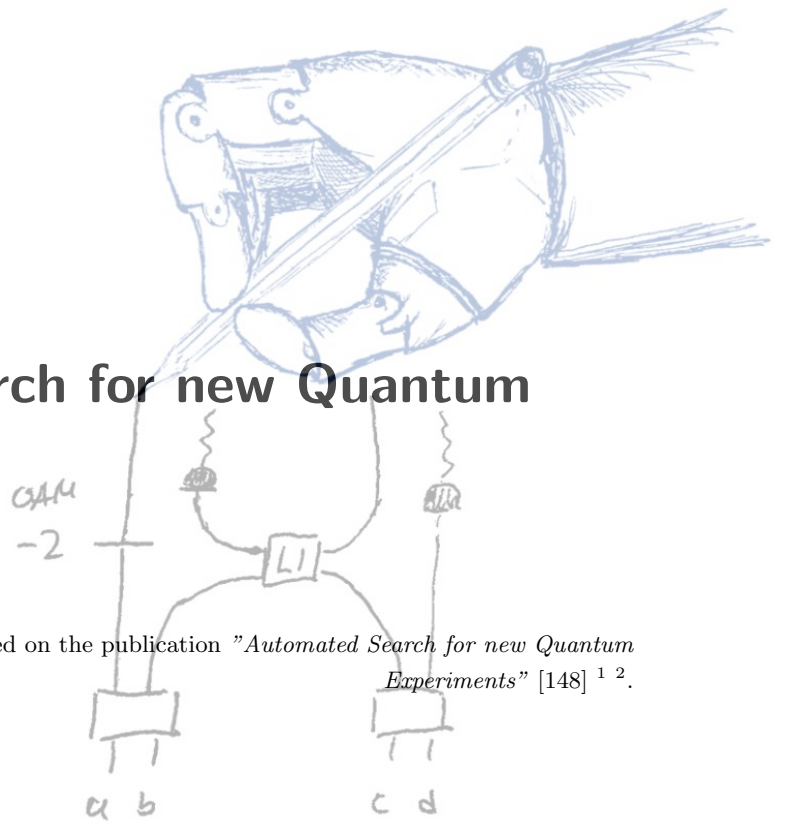
Here, the OAM degree-of-freedom is advantageous because, on the one side, it can is well known how to create and measure entangled states in OAM (in contrast to for instance frequency/time-bin encoding, where measurements are very challenging [154]), on the other side, they do not require interferometric stability (such as path-encoding [71, 155]). This allows to transmit (quantum) information encoded in OAM over large distances (so called *flying qubits*, or rather, *flying quDits* – as we have discussed in chapter 4).

The only remaining challenge seems to be the design of the actual experimental setup, but one would expect that that inspirations from two-dimensional multi-photon entanglement experiment [59, 95, 156] can be used to find high-dimensional generalisation.

However – it turned out that this expectation is not true, and when we started this project, we found ourselves in an awkward situation: We wanted to investigate the first high-dimensional generalisation of a Greenberger-Horne-Zeilinger state, but found it very challenging to find corresponding experimental setups creating them. None of our hand-designed setups led to a feasible solution. After some weeks I automated the search for this implementation – and

¹Mario Krenn, Mehul Malik, Robert Fickler, Radek Lapkiewicz, Anton Zeilinger, Automated Search for new Quantum Experiments. *Phys. Rev. Lett.* **116**, 090405 (2016).

²The background shows a robot which designs an experimental setup for a 3-dimensional Greenberger-Horne-Zeilinger state (by Robert Fickler).



(surprisingly to me) was able to find it within one night of search. Here I explain the computer algorithm, which we call MELVIN, that was able to automatically search and identify a feasible experimental setup for the state in question. The algorithm found the experimental design we were looking for – a high-dimensional multipartite GHZ state – and many other examples of high-dimensional multi-photon entangled states. In addition, we were able to find new quantum transformations, namely high-dimensional cyclic operations (which is a high-dimensional X-gate [157]), which we were not successful to design by hand. As a third example, the outputs of the algorithm served as inspiration for the development of very flexible new sources of quantum entanglement based on the concept of path identity. This was not achieved by searching for specific implementations, but by analysing the techniques of a specific outlier. Several of the computer designed experiments have been successfully implemented in our laboratories [98, 158], others are in preparation at the moment.

5.2 The algorithm

The main goal is to develop an algorithm which finds experimental implementations for certain interesting quantum states or quantum transformations. The basic idea is depicted in Figure (5.1). Initially, specific input states and a toolbox of experimentally known transformations, that will be used by MELVIN (the name of our algorithm), are defined. Using the elements from the toolbox, the algorithm assembles new experiments by arranging elements randomly. Then, from the initial state the resulting quantum state and transformation is calculated and its properties are analyzed. Well-defined criteria that are provided by the user decide whether the calculated quantum state has the desired properties. If the quantum state’s properties satisfy the criteria, the experimental configuration is simplified and reported to the user. MELVIN can store the configuration in order to use it as a basic building element in subsequent trials. Extending the initial toolbox (i.e. learning from experience) significantly speeds up the discoveries of more complex solutions.

Our method aims to create and manipulate general complex quantum states for which experimental implementations of arbitrary transformations are not known. The algorithm creates experiments using only experimentally accessible optical components, and selection criteria can involve properties that have practical advantages in the implementation. In the related but different field called computer-assisted or automated quantum circuit synthesis (QCS) [159–163], optimal implementations for quantum algorithms are constructed from universal sets of known quantum gates. The technique of QCS is used for linear qubit networks and usually requires fault-tolerant quantum computers for the implementation of its results (which is also called *compiler* [164]). In contrast to that, our method does not require a universal set of quantum gates because we focus on experimental implementable results – thus MELVIN’s toolbox only contains elements that we have in our laboratories. Furthermore, MELVIN can use multiple, potentially high-dimensional, degrees of freedom, as well as non-linear interactions (which can not be described in linear quantum networks). Furthermore, it can use different types of input states such as single photons, laser light, thermal light. Since MELVIN respects those experimental possibilities and constraints, several of these computer-designed experimental proposals have already been successfully implemented in our laboratories – several others are being built

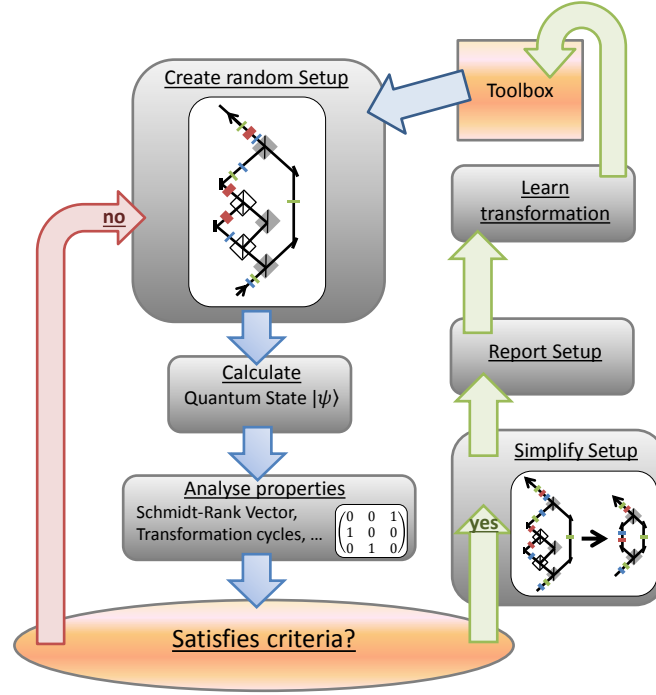


Figure 5.1: Working principle of our algorithm MELVIN. First, an experimental proposal is created using elements from a basic toolbox. Then, the quantum state is calculated, and subsequently its properties are analyzed. Those properties are compared with a number of criteria. If these criteria are not satisfied, the algorithm starts over again. However, if the criteria are satisfied, the experimental proposal is simplified and reported, together with all relevant information for the user. Useful solutions can be stored and used in future experimental configurations, which significantly increases the discovery rate of more complex experiments. The orange boxes (toolbox and criteria) are adapted when a different property of the quantum state or the quantum transformation is analysed by MELVIN, while the rest of the algorithm stays the same.

at the moment.

5.2.1 Symbolic description language

All quantum states are calculated using symbolic algebra using Wolfram Mathematica. A single-photon state is denoted as

$$\mathbf{a}[p_1, p_2, \dots, p_n] \quad (5.1)$$

where $\mathbf{a}$ indicates the path of the photon, and p_i are the properties of different degrees of freedom. For example,

$$\psi = \mathbf{a}[\ell = +2] \cdot \mathbf{b}[\ell = -2] \quad (5.2)$$

describes a state with two photons, one in path $\mathbf{a}$ and one in path $\mathbf{b}$ and ℓ is the OAM of the photon, which is $\ell = +2$ and $\ell = -2$. Every experimental element is calculated as a symbolic

modification of the input state. As an example, a 50/50 symmetric non-polarizing beam splitter for photons (only considering the OAM degree-of-freedom) can be described by

$$\text{BS}[\psi, a, b] = \psi \Leftarrow \begin{cases} a[\ell] \rightarrow \frac{1}{\sqrt{2}} (b[\ell] + i \cdot a[-\ell]) \\ b[\ell] \rightarrow \frac{1}{\sqrt{2}} (a[\ell] + i \cdot b[-\ell]) \end{cases} \quad (5.3)$$

where $\Leftarrow$ stands for a symbolic replacement followed by a list of substitution rules. ℓ stands for the orbital angular momentum (OAM) quantum number of the photon, and a and b denote the input paths of the beam splitter. Here, for simplicity, all other degrees of freedom (such as polarization or frequency) are considered to be the same for all photons. A very simple example is the operation of the beam splitter in equation (5.3) on the state in equation (5.2). A sketch of the setup can be seen in figure 5.2. In the symbolic language it is written as

$$\begin{aligned} \text{BS}[\psi, a, b] &\rightarrow \left(\frac{i \cdot a[-2] + b[2]}{\sqrt{2}} \right) \cdot \left(\frac{a[-2] + i \cdot b[2]}{\sqrt{2}} \right) \\ &\rightarrow i \left(\frac{a[-2]^2 + b[2]^2}{2} \right) \end{aligned} \quad (5.4)$$

which shows that the two photons leave the beam splitter always in the same arm. That effect is known as Hong-Ou-Mandel interference [165].

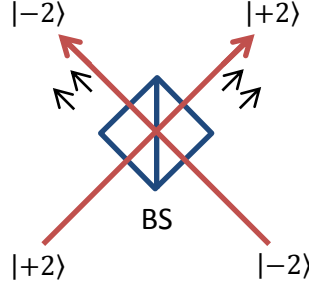


Figure 5.2: A simple sketch of a two-photon Hong-Ou-Mandel effect, to illustrate the example in equation (5.4). Two photons with opposite OAM arrive at a beam splitter, which leads to two photons either in the left or in the right output.

By realizing the calculations with symbolic algebra (instead of, for instance, linear algebra), adding new elements or even new degrees of freedom is very easy. Furthermore, it allows an intermediate forms which can easily be read by humans, important for the examination of solutions and the novel techniques found by the algorithm (which will be shown in chapter 5.6).

5.2.2 Experimental components

In the next step, I explain the elements which are used by the algorithm. This involves in particular the source of two-photon states, and elements which can modify the states – all of which are available in the laboratory. Many other elements can be added quite easily, just by specifying their precise action on the quantum state.

Spontaneous parametric down-conversion

Spontaneous parametric down-conversion (SPDC) is a prominent source for creating photon pairs. In one of the examples below, this non-linear process is used in order to create multi-photon entangled states. OAM is conserved in the SPDC process [3], $\ell_p = \ell_A + \ell_B$. Here we consider only pump beams with zero OAM, i.e. $\ell_p = 0$, which means the two photons will always have opposite OAM (in the paraxial approximation). For weak pump lasers, one can approximate the generated state by

$$|\psi_{SPDC}\rangle = \left(\sum_{\ell=-DC}^{DC} |+\ell_A, -\ell_B\rangle \right) \quad (5.5)$$

where DC is the order of down-conversion considered. For example, for $DC = 1$, only modes up to $\ell = 1$ are calculated. In the experiment, higher order modes usually have smaller probability (for instance, see [91]), therefore it is often sufficient to consider only low-order modes.

In the symbolic notation, a very similar notation can be used,

$$SPDC[\psi, a, b, DC] = \psi + \sum_{\ell=-DC}^{DC} a[+\ell]b[-\ell] \quad (5.6)$$

The creation of an initial double-pair in two different crystals can simply be written as

$$\begin{aligned} SPDC[0, a, b, 1] \cdot SPDC[0, c, d, 1] &= \\ &= (a[-1]b[+1] + a[0]b[0] + a[+1]b[-1]) \cdot (c[-1]d[+1] + c[0]d[0] + c[+1]d[-1]) \\ &= a[-1]b[+1]c[-1]d[+1] + a[0]b[0]c[-1]d[+1] + a[+1]b[-1]c[-1]d[+1] \\ &+ a[-1]b[+1]c[0]d[0] + a[0]b[0]c[0]d[0] + a[+1]b[-1]c[0]d[0] \\ &+ a[-1]b[+1]c[+1]d[-1] + a[0]b[0]c[+1]d[-1] + a[+1]b[-1]c[+1]d[-1] \end{aligned} \quad (5.7)$$

Here, double-pairs from one single crystal are neglected for simplicity. In the examples later in the text, those situations are considered as well – and will lead to interesting results. Polarisation properties can easily be included, but for simplicity they are not shown.

Reflection

A reflection of a mode inverts the OAM and gives a phase

$$\text{Reflection}[\psi, a] = \psi \Leftarrow a[\ell] \rightarrow i \cdot a[-\ell, H] \quad (5.8)$$

Non-polarizing symmetric 50/50 beam splitter

The beam splitter which also involves polarisation can be written as

$$BS[\psi, a, b] = \psi \Leftarrow \begin{cases} a[\ell, P] \rightarrow \frac{1}{\sqrt{2}} (b[\ell] + \text{Reflection}[a[\ell, P]]) \\ b[\ell, P] \rightarrow \frac{1}{\sqrt{2}} (a[\ell] + \text{Reflection}[b[\ell, P]]) \end{cases} \quad (5.9)$$

The reflection has results in a polarisation dependent phase which is accounted for in the function `Reflection`.

Polarizing beam splitter

The polarizing beam splitter has four different potential actions

$$\text{PBS}[\psi, \mathbf{a}, \mathbf{b}] = \psi \Leftarrow \begin{cases} \mathbf{a}[\ell, H] \rightarrow \mathbf{b}[\ell, H] \\ \mathbf{a}[\ell, V] \rightarrow i \cdot \mathbf{a}[-\ell, V] \\ \mathbf{b}[\ell, H] \rightarrow \mathbf{a}[\ell, H] \\ \mathbf{b}[\ell, V] \rightarrow i \cdot \mathbf{b}[-\ell, V] \end{cases} \quad (5.10)$$

Half-Wave Plate

A half-wave plate only acts on the polarisation degree of freedom (in this example, with an angle of 90°) as

$$\text{HWP}[\psi, \mathbf{a}] = \psi \Leftarrow \begin{cases} \mathbf{a}[\ell, H] \rightarrow \mathbf{a}[\ell, V] \\ \mathbf{a}[\ell, V] \rightarrow -\mathbf{a}[\ell, H] \end{cases} \quad (5.11)$$

OAM hologram

The action of a hologram can be written as

$$\text{OAMHolo}[\psi, \mathbf{a}, \mathbf{n}] = \psi \Leftarrow \{\mathbf{a}[\ell, P] \rightarrow \mathbf{a}[\ell + n, P]\} \quad (5.12)$$

where $n \in \mathbb{N}$ denotes the OAM charge that is added to the beam.

OAM hologram superposition

It is possible to add OAM in superpositions, which can be written as

$$\text{OAMHoloSP}[\psi, \mathbf{a}, \mathbf{n}] = \psi \Leftarrow \left\{ \mathbf{a}[\ell, P] \rightarrow \frac{1}{\sqrt{2}} (\mathbf{a}[\ell, P] + \mathbf{a}[\ell + n, P]) \right\} \quad (5.13)$$

where $n \in \mathbb{N}$ denotes the OAM charge that is added to the beam.

Dove Prism

One significant optical tool is a so called Dove prism. It is an element performing three reflections. The element can be rotated by an angle α . A photon with OAM ℓ observes an additional phase of $\exp(i \cdot \alpha \cdot \ell)$. We fix the possible rotations to integer fractions of π :

$$\text{DP}[\psi, \mathbf{a}, \mathbf{n}] = \psi \Leftarrow \{\mathbf{a}[\ell, P] \rightarrow e^{i \frac{\pi}{n} \ell} \text{Reflection}[\mathbf{a}[\ell, P]]\} \quad (5.14)$$

OAM Parity Sorter

A final, very significant device is a OAM parity sorter, which has been introduced by Leach et al. in [166] (therefore also called *Leach-interferometer*). It is a Mach-Zehnder interferometer with an additional Dove prism in one of the two arms. If $n=1$, even ℓ modes get no phase, however odd ℓ modes get a phase of π . Therefore they leave in opposite outputs of the interferometers and are sorted. One can write

$$\text{LI}[\psi, \mathbf{a}, \mathbf{b}] = \text{BS}[\text{Reflection}[\text{Reflection}[\text{DP}[\text{Reflection}[\text{BS}[\psi, \mathbf{a}, \mathbf{b}], \mathbf{a}], \mathbf{a}, 1], \mathbf{b}], \mathbf{b}], \mathbf{a}, \mathbf{b}] \quad (5.15)$$

$$(5.16)$$

5.2.3 Simplifying experiments

After an experiment setup is found, it is simplified as shown in figure 5.1. For that, three different hand-crafted methods are used. The first one removes elements from the experiment and calculates whether the resulting state or transformation is still performed the same way. Such simplifications could remove elements in paths that are not accessed. An example, in which it is necessary to remove multi elements at the same time, is the following: Four beam splitter after each other form two Mach-Zehnder interferometers. Those have no effect if the phases are set correctly, but can only be removed together.

In the second method, the algorithm tried to replace more complicated elements (such as LI, PBS or DP) by mirrors. This works in cases where only specific modes access the element (for instance, if only vertically polarized photons access a PBS).

A third method tries to simplify the path structure of the experiment by restructuring the paths. For example, if two PBS are used after each other, one output of the second PBS will never be used, thus the second PBS can be removed and one path can be removed completely. Those three methods are applied iteratively, until no simplification is possible anymore.

5.3 Example 1: High-dimensional multipartite entanglement

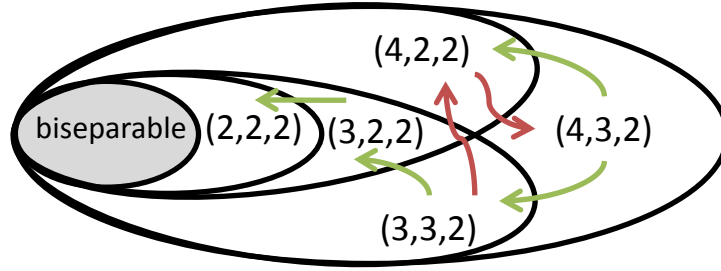


Figure 5.3: The structure of high-dimensional entanglement in a three-particle system in terms of Schmidt-Rank Vectors. The green/red arrows indicate allowed/forbidden transformations by using only local operations and classical communication (LOCC). The interpretation is, that the number of the SRV cannot be increased by local operations (and classical communication). Inspired by [149].

The Greenberger-Horne-Zeilinger (GHZ) state is the most prominent example for non-classical correlations between more than two involved parties, and has led to new understanding of the fundamental properties of quantum physics [58, 156]. It has been shown recently that its generalization to higher dimensions not only has curious properties [152], but that it is a limiting case of a much richer class of non-classical correlations [149, 150, 167]. Those new structures of multipartite high-dimensional entanglement are characterized by the *Schmidt-Rank Vector* (SRV) and give rise to new phenomena that only exist if both the number of particles and the number of dimensions are larger than two.

In the three-particle pure state case (which is the case we consider), the SRV represents the rank of the reduced density matrices of each party. If $|\psi\rangle$ is the quantum state of three particle

SRVs with known, computer-designed
experimental implementations

2,2,2	3,2,2	4,2,2	5,2,2	6,2,2	7,2,2	8,2,2	9,2,2	10,2,2	11,2,2	12,2,2
3,3,2	4,3,2	5,3,2	6,3,2	7,3,2	8,3,2	9,3,2	10,3,2	11,3,2	12,3,2	
3,3,3	4,3,3	5,3,3	6,3,3	7,3,3	8,3,3	9,3,3	10,3,3	11,3,3	12,3,3	
4,4,2	5,4,2	6,4,2	7,4,2	8,4,2	9,4,2	10,4,2	11,4,2	12,4,2		
4,4,3	5,4,3	6,4,3	7,4,3	8,4,3	9,4,3	10,4,3	11,4,3	12,4,3		
4,4,4	5,4,4	6,4,4	7,4,4	8,4,4	9,4,4	10,4,4	11,4,4	12,4,4		
	5,5,2	6,5,2	7,5,2	8,5,2	9,5,2	10,5,2	11,5,2	12,5,2		
	5,5,3	6,5,3	7,5,3	8,5,3	9,5,3	10,5,3	11,5,3	12,5,3		
	5,5,4	6,5,4	7,5,4	8,5,4	9,5,4	10,5,4	11,5,4	12,5,4		
	5,5,5	6,5,5	7,5,5	8,5,5	9,5,5	10,5,5	11,5,5	12,5,5		
	6,6,2	7,6,2	8,6,2	9,6,2	10,6,2	11,6,2	12,6,2			
	6,6,3	7,6,3	8,6,3	9,6,3	10,6,3	11,6,3	12,6,3			
	6,6,4	7,6,4	8,6,4	9,6,4	10,6,4	11,6,4	12,6,4			
	6,6,5	7,6,5	8,6,5	9,6,5	10,6,5	11,6,5	12,6,5			
	6,6,6	7,6,6	8,6,6	9,6,6	10,6,6	11,6,6	12,6,6			
	7,7,2	8,7,2	9,7,2	10,7,2	11,7,2	12,7,2				
	7,7,3	8,7,3	9,7,3	10,7,3	11,7,3	12,7,3				
	7,7,4	8,7,4	9,7,4	10,7,4	11,7,4	12,7,4				
	7,7,5	8,7,5	9,7,5	10,7,5	11,7,5	12,7,5				
	7,7,6	8,7,6	9,7,6	10,7,6	11,7,6	12,7,6				
	7,7,7	8,7,7	9,7,7	10,7,7	11,7,7	12,7,7				
		8,8,2	9,8,2	10,8,2	11,8,2	12,8,2				
		8,8,3	9,8,3	10,8,3	11,8,3	12,8,3				
		8,8,4	9,8,4	10,8,4	11,8,4	12,8,4				
		8,8,5	9,8,5	10,8,5	11,8,5	12,8,5				
		8,8,6	9,8,6	10,8,6	11,8,6	12,8,6				
		8,8,7	9,8,7	10,8,7	11,8,7	12,8,7				
		8,8,8	9,8,8	10,8,8	11,8,8	12,8,8				
			9,9,2	10,9,2	11,9,2	12,9,2				
			9,9,3	10,9,3	11,9,3	12,9,3				
			9,9,4	10,9,4	11,9,4	12,9,4				
			9,9,5	10,9,5	11,9,5	12,9,5				
			9,9,6	10,9,6	11,9,6	12,9,6				
			9,9,7	10,9,7	11,9,7	12,9,7				
			9,9,8	10,9,8	11,9,8	12,9,8				
			9,9,9	10,9,9	11,9,9	12,9,9				
				10,10,2	11,10,2	12,10,2				

not possible
DC = 1
DC = 2
DC = 3

Figure 5.4: Experimental implementations for high-dimensional three-partite entangled states proposed by MELVIN. The list shows experimental setups with states of different Schmidt-Rank Vectors. Black cells are not possible for combinatorial reasons [149]. Cells with green, violet and blue filling indicate states require a pair of 3-, 5-, 7-dimensional two-photon states to begin with. For white cells, no experimental realization has been found yet. It is not known why no experimental designs have been found for states in white cells. Either the search was not exhaustive enough, or they cannot be created with the provided elements in the toolbox. Furthermore, the apparent structure of green/violet states may indicate that the SRV is directly connected with the probability for the existence of experimental designs – but the reason for this is not known either.

A, B and C, then the ranks of the reduced density matrices

$$\begin{aligned}
 r_A &= \text{rank}(\text{Tr}_B(|\psi\rangle\langle\psi|)), \\
 r_B &= \text{rank}(\text{Tr}_A(|\psi\rangle\langle\psi|)), \\
 r_C &= \text{rank}(\text{Tr}_C(|\psi\rangle\langle\psi|))
 \end{aligned} \tag{5.17}$$

together form the SRV $d_\psi = (r_A, r_B, r_C)$ (for non-increasing order of r_i). The interpretation is the dimensionality of entanglement (Schmidt-Rank) between every part of the state with the remaining part (such as A—BC, B—AC, C—AB - which gives the entanglement between A with BC, B with AC and C with AB). It can also be understood now that the r_i can not be increased with local operations and classical communication. There are very instructive states which show the meaning of this quantity. One of such examples with Schmidt-Rank Vector

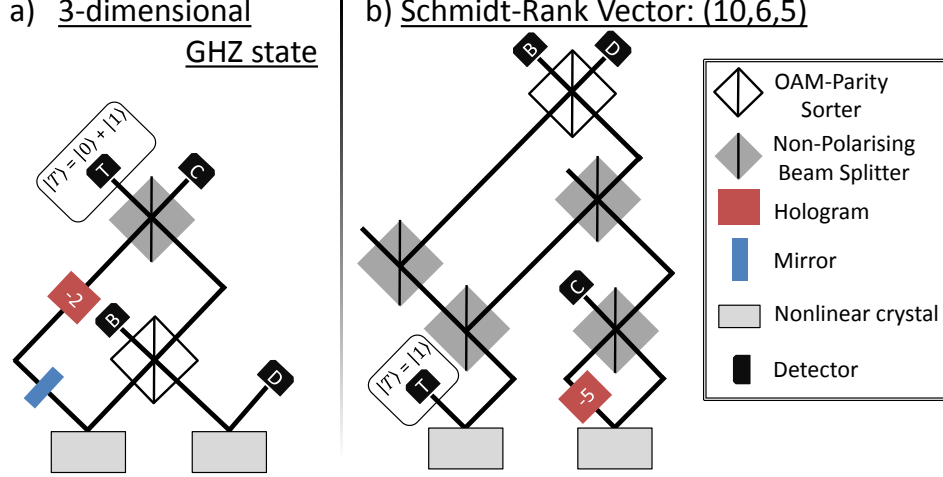


Figure 5.5: Experimental implementations of high-dimensional multipartite entangled quantum states. A) The experimental implementation for a 3-dimensional 3-partite GHZ-State. If Detector T (Trigger) observes a photon in the state $|\psi\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |1\rangle)$, then the rest of the quantum state is in a GHZ-state, which looks like $|\psi\rangle = \frac{1}{\sqrt{3}}(|0,0,0\rangle + |1,1,1\rangle + |2,2,2\rangle)$ (up to local transformations). The parity sorter, as described in [166], can sort even and odd OAM modes. The 3-dimensional GHZ-state has a Schmidt-Rank vector of (3,3,3) (all components are symmetrically entangled with the rest of the state). In this abstract representation, all elements are assumed to be transmissive, including the mirror. The experimental implementation is of course much more involved. B) A more complex experimental setup is required for higher-order Schmidt-Rank vectors. The (10,6,5)-state is one example of asymmetrically entangled quantum states. The experimental setups are just two examples of 51 implementations found for creating a variety of different entangled states, see figure 5.4.

(4,2,2) is the asymmetrically entangled state

$$|\psi\rangle = \frac{1}{2}(|0,0,0\rangle + |1,0,1\rangle + |2,1,0\rangle + |3,1,1\rangle). \quad (5.18)$$

There, the first particle is 4-dimensionally entangled with the other two parties, whereas particle 2 and 3 are both only two-dimensionally entangled with the rest. This Master-Slave-Slave configuration is one of the yet unexplored features that only exist in genuine high-dimensional multipartite entanglement, and will be interesting to study in more detail in future - both for fundamental properties as well as for their applications in novel quantum communication protocols [98]. A graphical representation of the three-particle structure in terms of SRVs can be seen in figure 5.3.

In order to make future experimental investigations possible, we aim to find high-dimensional multipartite entangled states in photonic systems. Here, the initial state is created by a double spontaneous parametric down-conversion process (SPDC). SPDC is a widespread source for experimental generation of photon pairs. Multiple SPDC processes can produce multipartite entanglement, as it is well-known for the case of two-dimensional polarization entanglement [59, 156]. However, instead of polarization, we use the orbital angular momentum

5 Automated Search for new Quantum Experiments

(OAM) of photons, which is a discrete high-dimensional degree of freedom based on the spatial structure of the photonic wave function, as explained in chapter 2.2.2.

The experimental designs are generated using a set of basic elements consisting of beam splitters, mirrors, Dove prism, holograms and OAM-parity sorters [166]. The holograms and the Dove prisms have discrete parameters corresponding to the OAM and phase added to the beam, respectively. These elements are randomly placed in one out of six different paths (four of the paths are inputs of the two photon pairs and two are empty to increase variability). One arm is used to trigger the tripartite state in the other three arms, which leads to roughly 10^{15} possible configurations. At the end, a postselection procedure consisting of the coincidence detection of four photons in the first four arms yields the final state. We calculate the Schmidt-Rank-Vector of the final state and select non-trivial ones (i.e. where there are no separable parties). Furthermore, for higher usefulness in experiments, we demand that the final state is maximally entangled in its orbital-angular momentum. If the criteria hold, the experimental setup is reported.

MELVIN runs for roughly 150 hours (on an Intel Core i7 notebook with 2,4GHz and 24 GB RAM), and finds 51 experiments for states that are entangled in genuinely different ways (see figure 5.4). Among them, we find the first experimentally realizable scheme of a high-dimensional GHZ-state [152], a generalization of the well-studied two-dimensional GHZ-state (see figure 5.5A). Furthermore, we find many experiments for different asymmetrically entangled states (such as the (4,2,2)-state explained above). In addition, several experiments only differ by continuously tunable components (e.g. different holograms or triggers), making it possible to explore continuous transitions between states of different classes of entanglement.

The resulting experiments contain interesting novel experimental techniques previously unknown – at least to the author. For example, in 50 out of 51 experiments, one of the four paths that comes directly from the crystals has not been mixed with any other arm (arm D in figure 5.5A, and arm T in figure 5.5B). The reason is that for double SPDC events it is possible that the two photon pairs come from the same crystal. Leaving one path unmixed leads to erasure of such double-pair emission events in four-fold coincidence detection. Interestingly, this immediately introduces asymmetry in the final experimental configuration. A different novelty is introduced when more than 6-dimensional entanglement is created beginning from two three-dimensional entangled pairs. This is only possible when the OAM of the photons from two crystals are shifted with respect to each other, and combined at a beam splitter (a preliminary stage of the technique can be seen in figure 5.5A, where the OAM of the photon in arm C is shifted in order to reach a 10-dimensional output). In other experimental designs, the normalization of the state has to be adjusted in order to get a maximally entangled output. As neutral-density filters were not part of the toolbox, MELVIN instead used beam splitters as a 50%-filter (for example, figure 5.5A).

5.3.1 Details for the 3-dimensional GHZ-state experiment

*** Next, we will explain the experiment for the 3-dimensional GHZ-state in figure 5.5A in more detail. It consists of an OAM-parity sorter, a mirror, a +2 Hologram and a beam splitter. (Note that for simplicity, we do not write normalization-constants)

After down-conversion (neglecting the double-emissions from one crystal as they will be

5 Automated Search for new Quantum Experiments

filtered in the four-fold coincidence detection in the end. We also neglecting higher-order OAM modes which are produced in SPDC with significantly lower probability), we can write

$$\begin{aligned}
|\psi\rangle &= (|0,0\rangle + |1,-1\rangle + |-1,1\rangle)^2 = \\
&= |0,0,0,0\rangle + |0,0,1,-1\rangle + |0,0,-1,1\rangle + \\
&+ |1,-1,0,0\rangle + |1,-1,1,-1\rangle + |1,-1,-1,1\rangle + \\
&+ |-1,1,0,0\rangle + |-1,1,1,-1\rangle + |-1,1,-1,1\rangle
\end{aligned} \tag{5.19}$$

After the OAM-Parity sorter (again, neglecting double-photons in one detector-arm, because they will be filtered by four-fold coincidence detection). As a result, only terms can lead to four-fold coincidences, which have the same OAM parity (parity= $\ell \bmod 2$):

$$|\psi\rangle = |0,0,0,0\rangle + |1,-1,1,-1\rangle + |1,-1,-1,1\rangle + |-1,1,1,-1\rangle + |-1,1,-1,1\rangle \tag{5.20}$$

For a Trigger in A with $(|0\rangle + |1\rangle)$, the SRV of the remaining state is SRV=(3,3,2), as one can see in the list above. Photon C and D reside in 3 dimensions while photon B lives in a 2-dimensional space. That is because photon B is perfectly anti-correlated with photon A, which is the 2-dimensional trigger. We want to increase the dimension of photon B to 3. The general idea is remove the perfect anti-correlation by mixing the trigger with photon C. At this stage, a BS between A and C would lead to Hong-Ou-Mandel interference between the 1st, 3rd and 4th term, which effectively removes those terms from the state and does not lead to a 3-dimensional GHZ state. In order to prevent this from happening, the OAM of the photon in A is shifted by -2. This prevents HOM interference between those three terms, and removes the 2nd term instead. One additional subtle but significant trick is the usage of a reflection in order to be not vulnerable to higher-order terms (without the mirror, the state would become a 2-dimensional GHZ if higher-order modes in SPDC are considered). The mirror in arm A leads to:

$$|\psi\rangle = |0,0,0,0\rangle + |-1,-1,1,-1\rangle + |-1,-1,-1,1\rangle + |1,1,1,-1\rangle + |1,1,-1,1\rangle \tag{5.21}$$

And the hologram of -2 in A transforms the state to

$$\begin{aligned}
|\psi\rangle &= |-2,0,0,0\rangle + |-3,-1,1,-1\rangle + |-3,-1,-1,1\rangle + |-1,1,1,-1\rangle + \\
&+ |-1,1,-1,1\rangle
\end{aligned} \tag{5.22}$$

In the next step, a beam splitter will be placed between arm A and C.

$$\begin{aligned}
|\psi\rangle &= |0,0,-2,0\rangle - |2,0,0,0\rangle + |1,-1,-3,-1\rangle - |3,-1,-1,-1\rangle + |-1,-1,-3,1\rangle - \\
&- |3,-1,1,1\rangle + \textcolor{red}{|1,1,-1,-1\rangle} - \textcolor{red}{|1,1,-1,-1\rangle} + |-1,1,-1,1\rangle - |1,1,1,1\rangle
\end{aligned} \tag{5.23}$$

The red terms cancel because of destructive interference. This is due to the Hong-Ou-Mandel effect, which occurs if the OAM of two incoming photons from two different arms in a beam splitter are opposite, which leaves us with the state

$$\begin{aligned}
|\psi\rangle &= |0,0,-2,0\rangle - |2,0,0,0\rangle + |1,-1,-3,-1\rangle - |3,-1,-1,-1\rangle + |-1,-1,-3,1\rangle - \\
&- |3,-1,1,1\rangle + |-1,1,-1,1\rangle - |1,1,1,1\rangle
\end{aligned} \tag{5.24}$$

If we now use the photon A as Trigger for $(|0\rangle + |1\rangle)$, the photons in B, C and D will be in the OAM state:

$$|\psi\rangle = |0, -2, 0\rangle + |-1, -3, -1\rangle - |1, 1, 1\rangle \quad (5.25)$$

As local unitary operation do not change the entanglement structure, the state is a variant of the three-dimensional three-particle GHZ state $|\psi\rangle = |0, 0, 0\rangle + |1, 1, 1\rangle + |2, 2, 2\rangle$. For that reason, the state in eq.(5.25) has the same physical properties as a GHZ state, even though it is encoded differently.

5.4 Example 2: High-dimensional cyclic operations and learning

In the second example, we are interested in high-dimensional cyclic rotations, which are special cases of high-dimensional unitary transformations. In particular, these transformations are generalizations of the Pauli σ_X gates into higher dimensions, and are important for building general unitary transformations [157]. A set of states is transformed in such a way that the last element of the set transforms to the first element (for example, $|1\rangle \rightarrow |2\rangle \rightarrow |3\rangle \rightarrow |1\rangle$ is a 3-cycle). Such transformations are required in novel kinds of high-dimensional quantum information protocols [168, 169] as well as in the creation of high-dimensional Bell-states. Here, our input is a set of high-dimensional states encoded in different degrees of freedom (path, polarization, and OAM). While the creation and verification of high-dimensional entanglement in OAM is well known [57, 81], the knowledge of how to perform arbitrary transformations in this degree-of-freedom is still lacking. Thus, finding such transformations in OAM is important, as it would enable practical experimental setups with high-dimensional quantum states and find application in high-dimensional quantum information protocols.

The experimental designs are generated using a set of basic elements that consists of polarizing and non-polarizing beam splitters, Dove prisms, mirrors, holograms and half-wave plates. These elements are placed in one of three different paths (one path is used as an input, and two empty paths are added to increase variability). This leads to roughly 10^{22} different possible experimental configurations.

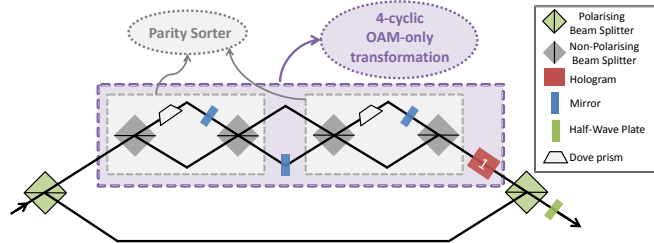


Figure 5.6: Realization of an 8-cyclic rotation using polarization and OAM ($| - 1, V\rangle \rightarrow | - 1, H\rangle \rightarrow |0, V\rangle \rightarrow \dots \rightarrow |2, H\rangle \rightarrow | - 1, V\rangle$). In the experiment, a 4-cyclic rotation for pure OAM values is used. Within the 4-cyclic rotation, the parity sorter [166] is used twice.

5 Automated Search for new Quantum Experiments

The criterion is based on the largest cycle of the transformation: A number of input states (with different polarization (horizontal and vertical), different OAM ($\ell=-10 \dots +10$) and different paths) is calculated. Then we search subsets of modes that are transformed in a closed cycle, as described above, and select the largest closed cycle. MELVIN was able to find the first experimentally realizable OAM-only 4-cyclic transformation, OAM and polarization 3-, 6-, and 8-cyclic rotations and up to 14-cyclic rotations using OAM, polarization and path (see figure 5.6).

5.4.1 Learning from previous Experimental Designs

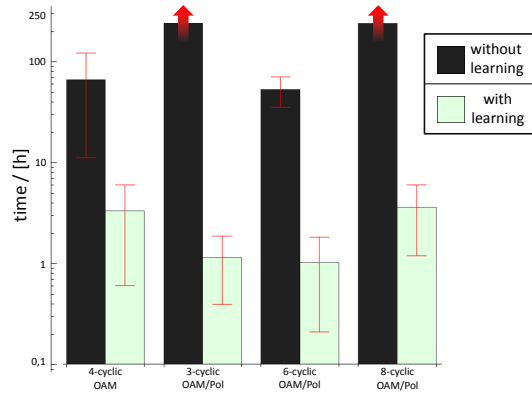


Figure 5.7: Comparison of performance with and without the ability to learn (log-scale). Green shows the average time required in the case where the algorithm can learn useful transformations (the algorithm was executed 10 times with the same initial conditions). Black shows the time it requires without the ability to learn. The experimental setup for 3-cyclic and 8-cyclic transformations have not found (within 250 hours) without learning, while experimental designs for 4-cyclic and 6-cyclic rotation were found three and four times in 250 hours, respectively. The errors stand for one standard deviation, calculated from the times it took to find the solution. Thus, autonomously extending the set of useful transformations improves MELVIN’s performance, which is crucial for scaling to more complex experimental designs.

Complex problems can be solved more efficiently by reusing solutions to simpler problems [170, 171]: Whenever MELVIN finds a solution for a simple system, it memorizes the experimental configuration as new part of its initial toolbox. The novel elements in the toolbox can be used to construct the next experimental configuration. The algorithm extends its own set of basic elements autonomously, based on the properties of the longest cycle. It saves elements that have large cycles and experimental setups with non-trivial coupling between different degrees-of-freedom. Additionally, elements already learned can be *forgotten* to improve variability and prevent dead ends, as some of them might even have negative effects on the probability of finding new experiments. The decision of which elements are forgotten at which times is purposefully random. Even though it would be possible to weight the elements for past usefulness, it would introduce a bias on similar solutions that we wanted to prevent.

To investigate the effectiveness of learning, we analyze the algorithm with and without the

ability to increase its own set of basic elements. We ran the algorithm for 250 hours, and only 3 and 4 instances of 4-cyclic and 6-cyclic rotations were found, respectively. Not a single instance of a 3-cyclic and an 8-cyclic rotation was found within 250 hours. However, using the ability to learn new elements we ran the algorithm 10 times (starting with the initial toolbox, i.e. without keeping the learned elements), and discovered that the 3- and 6-cyclic rotations were found on average within 90 minutes (they were always found within 3 hours), and the 4- and 8- cyclic rotations were found on average within 3,5 hours (in each of the 10 trials, they were found within 8 hours). Thus the ability to learn new elements improves the search by more than one order of magnitude, suggesting a mechanism for experiments with a higher complexity (see figure 5.7).

In the end, the 4-cyclic cyclic rotations (fully within the OAM degree of freedom) found by the algorithm is described, the other solutions are shown in the Appendix A. ψ stands for the initial state and XXX stand for the state after the previous element.

4-cyclic OAM-only rotation

Experimental configuration:

$$\begin{aligned}
 \text{BS}[\psi, a, b] &\rightarrow \text{DP}[XXX, b, 1] \rightarrow \text{Reflection}[XXX, b] \rightarrow \text{BS}[XXX, a, b] \\
 &\rightarrow \text{Reflection}[XXX, a] \rightarrow \text{BS}[XXX, a, b] \rightarrow \text{DP}[XXX, b, 1] \\
 &\rightarrow \text{Reflection}[XXX, b] \rightarrow \text{BS}[XXX, a, b] \rightarrow \text{OAMHolo}[XXX, a, 1]
 \end{aligned}
 \tag{5.26}$$

Operation:

$$|-1\rangle \rightarrow |0\rangle \rightarrow |1\rangle \rightarrow |2\rangle \rightarrow |-1\rangle \tag{5.27}$$

The number in the ket stands for the OAM. This experiment has been performed in our laboratories for classical laser beams [158] and on the single-photon level [157].

5.5 Experimental implementations

For implementing the computer-designed setups, several experimental requirements and restrictions need to be taken into account. Multi-Photon experiments are usually performed using pulsed lasers. The probability p that a laser pulse generates a photon pair in an SPDC process is essential here. On the one hand, for a 4-photon experiment, p^2 must be large enough to obtain a large rate of . On the other hand, p^3 must be small enough to produce as few as possible unwanted triple pairs (*accidental counts*), because they are one of the main reasons for reduced quality of the generated state [95, 172]. As a tradeoff, for many multi-photon (polarisation-entanglement) experiments p is in the order of 10%.

The transformation- and detection efficiencies is another important factor which distinguishes the computer-designed proposals with the actual experiments. The detection efficiencies D of commonly used single-photon detectors is roughly $D=75\%$ – for four-photon experiments this results in the detection of roughly one in three quadruples. An obvious (but expensive) way to improve this is usage of nanowire detectors. The overall efficiencies of Spatial Light

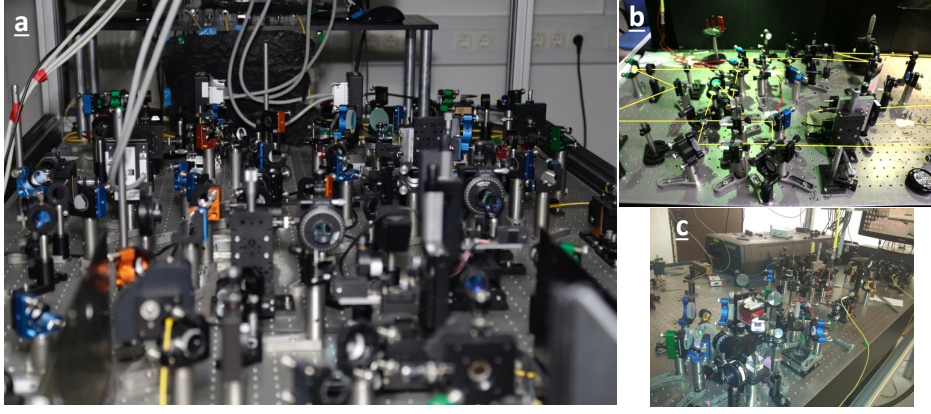


Figure 5.8: Photos of experimental successful implementations of MELVIN’s proposals. **a:** The experimental implementation of a high-dimensional multi-photon entangled state. (Image by Manuel Erhard) **b:** Implementation of the 4-dimensional cyclic transformation with lasers [158]. (Photo by Florian Schlederer) **c:** Implementation of the 4-dimensional cyclic transformation on a single-photon level [157] (Photo by Feiran Wang)

Modulators is also roughly 75% (which is due to losses in the reflection, and due to imperfect gratings), which – for four-photon experiments – leads to another factor of $0.75^4 = 0.3$ in detected photon four-folds.

In multi-photonic entanglement experiments, the difference of the group velocity between the pump beam and the down-converted photons is important. Intuitively, the pump photons and the down-converted photons have a different velocity in the crystal. Thus when the SPDC process happens in the beginning of the crystal, the time difference between pump and down-conversion photons is large. When the process happens in the end of the crystal, the difference is small. This leads to an inherent timing jitter. When photons are created in two crystals, there must not be (in principle) information to distinguish them - in particular by their arrival time. If the coherence time of the photons is smaller than the timing jitter, one could distinguish them. For this reason, the spectral bandwidth of the photons need to be filtered to increase the coherence time [173].

High-dimensional entangled states in the OAM basis have the form of $|\psi\rangle = N \cdot \sum_{\ell=0} a_{|\ell|} |\ell, -\ell\rangle$ [3]. The coefficients $a_{|\ell|}$ decrease when ℓ increases, thus the states are not maximally entangled (for instance, see [57, 81, 174]). For multi-photon entanglement experiments, these coefficients lead to non-maximally entangled multi-photon states. In some special cases (such as for the experiment in [98]), a non-equal trigger can be used to get maximally entangled multi-photon states.

Many of the proposed experiments involve interferometers (such as the parity sorter [166]). Interferometers need to be stable below the wavelength of the photons. The experiment demonstrating the cyclic operation with lasers [158] involves two parity sorter. The visibility of the interferometer dropped from 95% to 75% within 60 minutes [175]. In high-dimensional multi-photon experiment, data accumulation often takes much longer than one hour (for example, in [98], photon counts have been accumulated over 3 days). In this case, active stabilisation of

5 Automated Search for new Quantum Experiments

the interferometers are necessary, for instance via periodic test measurements and automated adjustments with piezo crystals.

5.6 Example 3: Entanglement by Path Identity

The content of this chapter is based on the publication "*Entanglement by Path Identity*" [176]³.

5.6.1 Inspired by a computer algorithm

In the first two examples, we used well-defined selection criteria which led to different experimental solutions with specific properties. In the following example, we obtained the idea (which we denoted *entanglement by path identity*) not by defining selection criteria, but by investigating unexpected results.

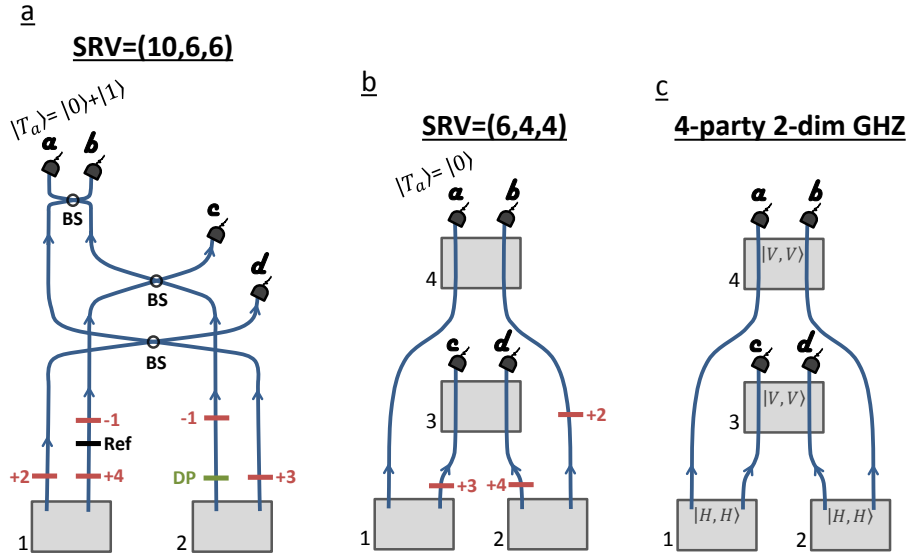


Figure 5.9: **a:** We investigated an experiment with an unexpectedly large Schmidt-Rank Vector (10,6,6). The experiment works because the crystals are producing four photons in a coherent superposition of one SPDC event in each of the crystals simultaneously, and a double-SPDC events in each of the crystals. Here, mode shifters (such as holograms) are shown in red, Dove prisms are green, BS stand for beam splitter. The trigger is in path *a*. **b:** By explicitly allowing superposition photon pairs being created in different crystals, and enabling overlapping of their output paths (but removing any other interaction such as beam splitters), the program was still able to find high-dimensional multi-photon experimental setups. **c:** Restricting the program further to allow only the two-dimensional polarisation entanglement and no trigger, it still found a solution. From that solution we understood the very basic idea, which we were able to generalize much further.

In particular, we let the program run for several weeks with the criteria from example 1 (maximally entangled states with a non-trivial Schmidt-Rank vector, in chapter 5.3). From there, we got a list of different accessible SRVs. One of the solutions was particularly unexpected: It is possible to create a SRV=(10,6,6) state starting only from two crystals with

³Mario Krenn, Armin Hochrainer, Mayukh Lahiri, Anton Zeilinger, Entanglement by Path Identity. *Phys. Rev. Lett* **118**, 080401 (2017).

three-dimensional entanglement. The experimental setup (depicted in figure 5.9a) works because it explicitly uses a superposition of SPDC happening twice in each individual crystal, and twice in crystal 1 and 2 together ($\text{SPDC}_1 \text{ SPDC}_1 + \text{SPDC}_1 \text{ SPDC}_2 + \text{SPDC}_2 \text{ SPDC}_1 + \text{SPDC}_2 \text{ SPDC}_2$). Superposition of different processes has been investigated in the context of induced coherence by Zou, Wang and Mandel in 1991 [177, 178]. In addition, in their investigation, the authors overlap the output paths of the crystals. For that reason, we added that possibility to MELVIN. As compensation, we removed the possibility to use beam splitters. Surprisingly, the program was still able to find non-trivial SRVs, as shown in figure 5.9b. In a final step, we restricted the program to the simplest situation of two-dimensional polarisation, not allowing for any triggers, and no entanglement produced in the crystals. Again MELVIN found a solution for a four-particle GHZ state entanglement (figure 5.9c). From that experiment, we understood the main idea of the new technique – and were able to generalize it to various different situations. It shows that automated designs of quantum optical experiments by algorithms can not only produce specific quantum states or transformations, but can also be a source for inspiration for new techniques – which can further be investigated by human scientists in more general contexts. This can be seen as a successful human-machine synergy.

5.6.2 Path identity

As mentioned above, Zou, Wang and Mandel reported in 1991 an experiment where they induce coherence between two photonic beams without interacting with any of them [177, 178]. They used two SPDC (spontaneous parametric down-conversion) crystals, where one photon pair is in a superposition of being created in crystal 1 and crystal 2 – which can be described as $|\psi\rangle = \frac{1}{\sqrt{2}}(|a\rangle|b\rangle + |c\rangle|d\rangle)$. The striking idea (originally proposed by Zhe-Yu Ou) was to overlap one of the paths from each crystal (Figure 5.10), which can be written as $|b\rangle = |d\rangle$. This method removes the *which-crystal information* of the final photon in path d . In contrast to a quantum eraser, the information here is not erased by postselection. Instead, all photons arrive in the same output irrespectively in which crystal they are created. The resulting state is to $|\psi\rangle = \frac{1}{\sqrt{2}}(|a\rangle + |c\rangle)|d\rangle$: One photon is in path d , and its partner photon is in a superposition of being in path a and c . There has been some follow-up work in recent years in the areas of quantum spectroscopy [179, 180], quantum imaging [181, 182], studies of complementarity [183–186], optical polarisation [187] and in microwave superconducting cavities [188]. However, this striking idea has not been investigated in the context of creating quantum entanglement yet.

The technique was first used by our program MELVIN, as shown in figure 5.9, and from there we generalized it: We show that by superposing photon pairs created in different crystals, and overlapping the photons paths, one can generate very flexible experiments producing various types of entanglement both in the multi-photon and the high-dimensional regime. First, we explain different schemes to produce various multi-photon polarization-entangled states such as Greenberger-Horne-Zeilinger (GHZ) states [58] and W states [189, 190], and contrast them with traditional methods [191]. The method is then generalized to high-dimensional multi-photon entangled states (such as a 4-particle 3-dimensional GHZ state), for which only few special cases existed so far [98, 148] – see chapter 5.3. Furthermore, we present for the first time a method to create arbitrary high-dimensional two-photon entangled states, for example,

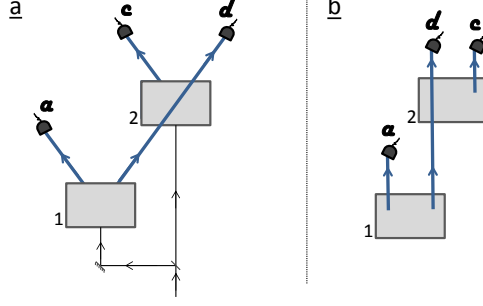


Figure 5.10: **a:** The simplest example which uses the overlapping modes has been discussed first in [177]. The two crystals (grey squares) can produce one pair of photons (blue lines), either in the first or in the second crystal with the pump beam depicted with black lines. If the two processes are coherent and the photons have same the frequency and polarisation, it is not known in which crystal the photons are created. In that case, the resulting photon pair is in the state $|\psi\rangle = \frac{1}{\sqrt{2}}(|a\rangle + |c\rangle)|d\rangle$. **b:** A simple sketch of the same experiment. For simplicity, we will use this more abstract representation of physical experiments in the rest of the text.

arbitrary high-dimensional Bell states in orbital angular momentum (OAM) or frequency of photons.

5.6.3 Multi-photon entanglement in Polarisation

First, we consider 4-photon polarization entanglement (Figure 5.11a), which was the output of MELVIN from where we understood the main idea. Crystal 1 and 2 can produce horizontally polarized pairs while crystal 3 and 4 can produce vertically polarized ones. The crystals are pumped coherently (for details of the coherence requirements, see section 5.6.4) and the pump power is adjusted such that we can neglect the cases where more than two pairs are created. The idea is that four-photon coincidences (i.e. one photon in each of the four paths) can only happen either when the two pairs come from crystal 1 & 2 or crystal 3 & 4. No other event produces four-photon coincidences. For example, if the pairs are produced in crystal 1 & 3, there will be two photons in path c , but none in path b . The resulting four-photon state can be written as

$$|\psi\rangle = \left(|H_a, H_c\rangle + |H_b, H_d\rangle + |V_a, V_b\rangle + |V_c, V_d\rangle\right)^2$$

$$|\psi\rangle \xrightarrow{4\text{-fold}} \frac{1}{\sqrt{2}} \left(|H_a, H_b, H_c, H_d\rangle + |V_a, V_b, V_c, V_d\rangle\right), \quad (5.28)$$

where H and V stand for horizontal and vertical polarisation, respectively, and the subscript stands for the photon's path. In an analogous way, by increasing the number of crystals and the pump power, entangled states with more photons can be created. In figure 5.11b the scheme for creating a n -photon GHZ state is shown.

In contrast to our new method, the traditional way of creating these states requires two crystals each producing a pair of polarization entangled photons. One photon from each crystal goes to a polarizing beam splitter (PBS), which removes the which-crystal information. Triggering on

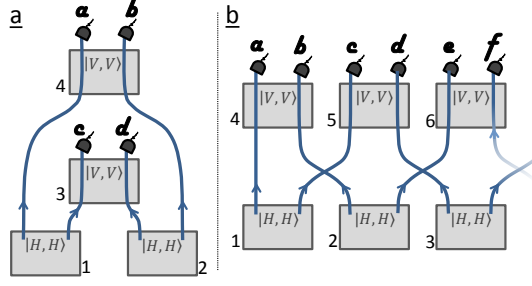


Figure 5.11: Multi-photon entanglement with polarization. **a:** In four crystals, two photon pairs are produced. Crystal 1 and 2 produce horizontally polarized photons, while crystal 3 and 4 produce vertically polarized ones. Four-photon coincidences can only happen when crystal 1 and 2 fire together or when crystal 3 and 4 fires. This leads to a 4-particle GHZ-state $|\psi\rangle = \frac{1}{\sqrt{2}} (|H, H, H, H\rangle + |V, V, V, V\rangle)$. **b:** Entangled states with more numbers of particles can be created in an analogous way – here a n -photon GHZ state $|\psi\rangle = \frac{1}{\sqrt{2}} (|H, H, H, \dots\rangle + |V, V, V, \dots\rangle)$ is shown.

events where all 4 detectors click, a 4-particle GHZ $|\psi\rangle = \frac{1}{\sqrt{2}} (|H, H, H, H\rangle + |V, V, V, V\rangle)$ state is created [192]. With that traditional method, GHZ-entanglement with 8 photons [59, 193] and very recently, up to 10 photons have been created [95, 96].

The new scheme does not need entangled photons to start with. Furthermore removing of the which-crystal-information using a PBS is not necessary. Our method is not taking advantage of cascaded down-conversion, as it has been down in recent articles producing multi-photon polarisation entangled states [194, 195].

In our examples, stimulated emission does not happen (which would introduce noise in the entangled state), because the input modes into the crystal are orthogonal to the output modes (for instance, having different polarization). In other cases, such as for the generation of the 4 photon W-state (figure 5.12), stimulated emission could happen but its contributions to the four-photon coincidences are neglectable.

5.6.4 Realistic experimental diagram and coherence time requirements

A possible implementation of the 4-photon GHZ state experiment described in Figure 5.11a, is shown in more detail in Figure 5.13. A laser beam (depicted in blue), which is split at a 50/50 beam splitter arrive at the same time at crystal 1 and 2. For that, the path length between the beam splitter and the crystals 1 and 2 (written as ℓ_{p1} and ℓ_{p2}) need are matched with a standard trombone system.

Crystals 1 and 2 can both produce photon pairs with horizontal polarization, while crystal 3 and 4 can both produce vertically polarized photon pairs – with non-collinear phase-matching conditions. This can be achieved, in analogy to the cross-crystal two-photon source, with type-II SPDC and a diagonally polarized pump beam.

In order to make sure that the states are produced in a coherent superposition, path lengths need to be chosen appropriately. In the example above (Figure 5.13), the condition that needs to be fulfilled that one cannot distinguish whether the 4-fold coincidence count came from crystal 1 and 2, or whether it came from crystal 3 and 4. For that, the path length $\ell_1 - \ell_4$ need to be

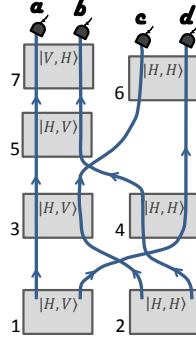


Figure 5.12: W-states such as $|\psi\rangle = \frac{1}{2}(|VHHH\rangle + |HVVH\rangle + |HHVH\rangle + |HHHV\rangle)$ represent a different type of multi-photon entanglement. While the GHZ state is considered as the most non-classical state, a W-state is the most robust entangled state because the loss of one particle leaves an entangled state. Here, four-folds can only happen if crystal 1 & 2 both produces a pair of photons, or crystal 3 & 4, or crystal 5 & 6 or 6 & 7. Interestingly, in this setup, one photon from crystal 1 can stimulate an emission in crystal 3. However, this will not lead to four-fold coincidences, as there is no photon in path d . Thus, this event can be neglected.

equal within the coherence length of the down-converted photons. Furthermore, the pump laser paths need to be matched, such that $\ell_{p1} = \ell_{p2}$ and $\ell_{p3} = \ell_{p4}$ within the coherence time of the laser. The final requirement is that the path lengths of the laser between the two crystals must be matched with the path length of the down-converted photons, such that $\ell_{p3} = \ell_1$ within the coherence time of the laser. It can be summarized as

$$|\ell_i - \ell_j| \ll c\tau_{SPDC}, \quad (5.29a)$$

$$|\ell_{p1} - \ell_{p2}| \ll c\tau_{pump}, |\ell_{p3} - \ell_{p4}| \ll c\tau_{pump}, \quad (5.29b)$$

$$|\ell_{p3} - \ell_i| \ll c\tau_{pump}, |\ell_{p4} - \ell_i| \ll c\tau_{pump}, \quad (5.29c)$$

where $c\tau_{SPDC}$ and $c\tau_{pump}$ are the coherence length of the down-converted photons and the pump laser, respectively. We find these restrictions on the temporal coherence and indistinguishability by applying the results that were obtained for the two-photon case by Jha et al. [196]. These are similar requirements as for standard 4-photon GHZ sources based on overlapping the photons at a polarizing beam splitter. Furthermore, also the requirements for the matching of frequencies is analogous to the standard 4-photon GHZ source using an PBS.

5.6.5 Multi-photon entanglement in higher dimensions

The principle can be generalized to produce high-dimensional multi-photon entangled states [149, 152]. As discussed above, high-dimensional entanglement has been investigated mainly in the two-photon case, with two recent exceptions which investigated three-dimensional entanglement with three photons [98], and teleportation of two degrees-of-freedom of a single photon [172]. Figure 5.14a shows our proposal for an experiment creating a 3-dimensional 4-party GHZ-state, starting from crystals which create separable photon pairs. There are 3 layers of 2 crystals – i.e. 6 crystals that are pumped coherently, and photon pairs are created in two

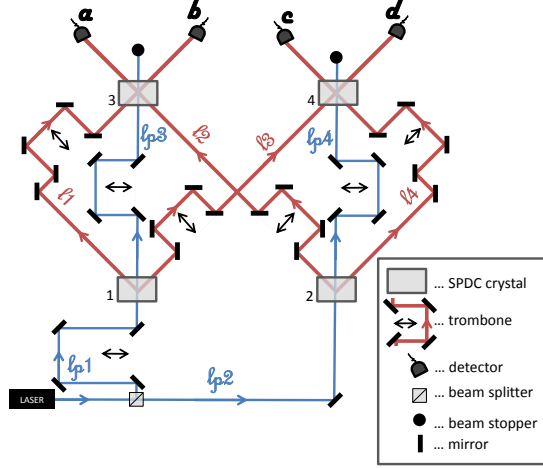


Figure 5.13: A realistic diagram of the experiment described in the main text, which produces a 4-photon GHZ-state in polarization. The pump path length ℓ_{p1} and ℓ_{p2} start at the BS and end at crystals 1 and 2. The path lengths ℓ_{p3} and ℓ_{p4} start at crystals 1 and 2, and end at crystals 3 and 4. The path lengths for the down-conversion photons ℓ_i start at the crystals 1 and 2, and end at crystals 3 and 4.

of them (because the pump power is set to such a level that higher-order emissions can be neglected).

Each photon from the first layer (crystals 1 and 2) passes through two mode shifters, which in total shift its mode by +2. In the case of orbital angular momentum (OAM) of photons, mode shifters are holograms which add one unit of OAM to the photon. Photons from the second layer (crystal 3 and 4) pass through one mode shifter, while the photons created in the uppermost layer (crystal 5 and 6) stay in their initial mode. The resulting 4-photon state can be described in the following way:

$$|\psi\rangle = \left(|2_a, 2_d\rangle + |2_b, 2_c\rangle + |1_a, 1_c\rangle + |1_b, 1_d\rangle + |0_a, 0_b\rangle + |0_c, 0_d\rangle \right)^2, \quad (5.30)$$

where 0, 1 and 2 stand for the mode number (such as the OAM of the photon), and subscript denotes the photon's path. In the same way as before, by neglecting cases where more than two photon pairs are produced, one finds that a 4-fold coincidence event in detectors a, b, c, d can only be created either if crystal 1&2 fire together or crystal 3&4 or crystal 5&6. This leads to

$$|\psi\rangle \xrightarrow{4\text{-fold}} \frac{1}{\sqrt{3}} \left(|2_a, 2_b, 2_c, 2_d\rangle + |1_a, 1_b, 1_c, 1_d\rangle + |0_a, 0_b, 0_c, 0_d\rangle \right). \quad (5.31)$$

Our idea can further be generalized to cases for more than 4 photons, to produce multi-photonic high-dimensional entangled states. Adding phase shifters between the crystals can be used to produce more general states. Adding additional columns increases the photon number n , while adding additional layers increases the dimensionality of entanglement d . For $n > 4$ and $d > 2$, the resulting quantum states are not in the form of GHZ states anymore. Analysing the general structure of produceable states opens a surprising connection to Graph Theory [197].

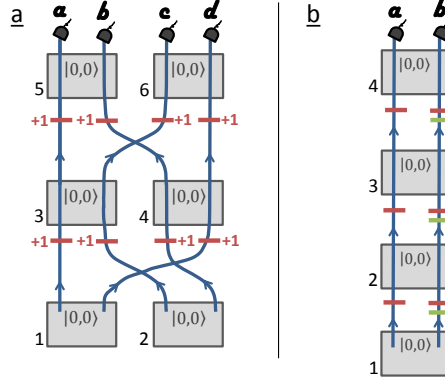


Figure 5.14: Multiphoton-Entanglement with high-dimensional degrees of freedom (in this example: orbital angular momentum). **a:** The setup produces two photon pairs. Four-Photon coincidences can only occur, when crystal 1 & 2 fire together, or crystal 3 & 4 or crystal 5 & 6. The produced photon pairs are all in the lowest mode (such as OAM=0). After each layer of crystals, a hologram increases the OAM of the photons (depicted as red line). After the third layer, photons from crystal 1 & 2 have OAM=2 and photons from the middle layer have OAM=1, which leads to a four-particle three-dimensional entangled GHZ-state ($|\psi\rangle = \frac{1}{\sqrt{3}}(|0,0,0,0\rangle + |1,1,1,1\rangle + |2,2,2,2\rangle)$). **b:** The same technique can also be applied to two-particle states to produce general high-dimensional states. All crystals produce pairs of Gaussian photons. The red lines indicate OAM holograms, the green lines indicate phase shifters. For example, if the holograms all are OAM=1 and phases are ignored, then the final output state is $|\psi\rangle = \frac{1}{2}(|0,0\rangle + |1,1\rangle + |2,2\rangle + |3,3\rangle)$. However, if one changes phases and the OAM in photon b, arbitrary 4-dimensional states can be created – for example all 16 four-dimensional Bell states. By increasing the number of crystals, more dimensions can be added.

5.6.6 Two-photon arbitrary high-dimensional entanglement

Finally, we show that the same technique can be applied to generate arbitrary high-dimensional entangled two-photon states, starting again with only separable (non-entangled) photon pairs. As shown in figure 5.14b, four crystals are set up in sequence (only one photon pair is produced) and their output modes are overlapped. Between each crystal, one adds arbitrary phase shifters and mode shifters (as before, holograms for OAM modes). That allows for adjusting every individual term in the superposition independently. For example, with all phases set to $\phi = \frac{\pi}{2}$ and all mode shifters being +1, the setup creates $|\psi\rangle = \frac{1}{2}(|0,0\rangle + i|1,1\rangle - |2,2\rangle - i|3,3\rangle)$. The dimension can be increased by increasing the number of layers (crystals); the minimum number of layers for creating a d -dimensional entangled state is d .

Traditional methods for producing high-dimensional entanglement (as that discussed in chapter 3) exploit the entanglement produced directly in a crystal. Such methods can only produce very restricted types of states. Furthermore, those states are never maximally entangled and have low rates of production. Our technique overcomes these restrictions and can produce arbitrary high-dimensionally entangled two-photon states. We can also tune the amount of entanglement in the following ways: 1) by adjusting the pump laser power between different

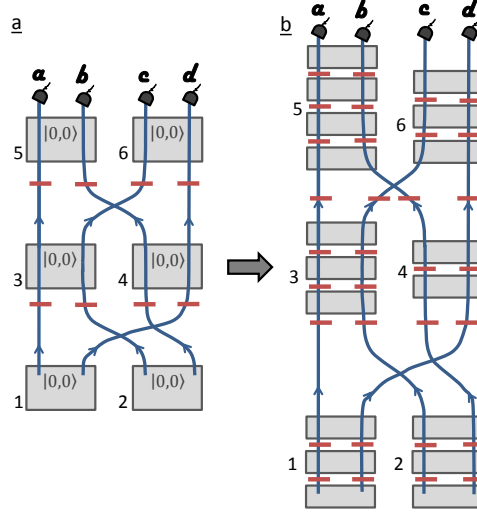


Figure 5.15: **a:** Experiment for an arbitrary 4-photon 3-dimensional GHZ state **b:** A generalisation of GHZ state. Every crystal can be replaced by a row of crystals each producing arbitrary high-dimensional 2-photon states. The red lines indicate an arbitrary mode shifter (such as holograms for OAM).

crystals, we can produce non-maximally entangled pure states; 2) by pumping the crystals with pumps that are not fully coherent to each other, we can produce entangled mixed states.

The number of created photon pairs does not depend on the number of crystals in the experimental setup. For example in figure 5.14b, even though there are 4 crystals, only one photon pair is created. Therefore the expected two-photon rate is of the same order as in conventional single-crystal source. Moreover, our method requires only separable photon pairs to begin with. Therefore, for the OAM of photons, the production rate can be significantly higher than the rate achievable with a traditional method (where higher-dimensional entanglement is created directly in the crystal). This is because it is substantially easier to create photon pairs in zero-order (Gaussian spatial mode) than in higher-order modes.

5.6.7 Construction of general experiments

For the case of four photons, the general constructable state can be written

$$\begin{aligned}
 |\psi_1\rangle = \frac{1}{N} & (|\ell_1, \ell_2, \ell_3, \ell_4\rangle + \\
 & c_{1,1} |\ell_5, \ell_6, \ell_7, \ell_8\rangle + \\
 & c_{1,2} |\ell_9, \ell_{10}, \ell_{11}, \ell_{12}\rangle)
 \end{aligned} \tag{5.32}$$

where $\ell_i \in \mathbb{Z}$ and $c_{i,j} \in \{0, e^{i\phi}\}$ (which is depicted in Figure 5.15a).

Each crystal could now be replaced by a row of crystals (such as Figure 5.14b), where each row produces an arbitrary 2-photon d-dimensional entangled states. That leads to the following

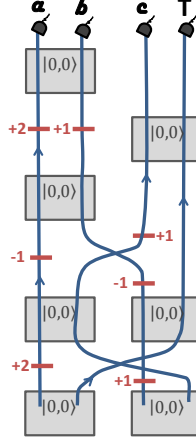


Figure 5.16: An experiment which creates an asymmetrically entangled quantum state, with a Schmidt-Rank-Vector of (4,2,2).

possible state

$$\begin{aligned}
 |\psi_2\rangle = \frac{1}{N} \big(& \\
 & \left(\sum_i c_{1,i} |\ell_{1,i}, \ell_{2,i}\rangle_{a,b} \right) \left(\sum_i c_{2,i} |\ell_{3,i}, \ell_{4,i}\rangle_{c,d} \right) + \\
 & \left(\sum_i c_{3,i} |\ell_{5,i}, \ell_{6,i}\rangle_{a,c} \right) \left(\sum_i c_{4,i} |\ell_{7,i}, \ell_{8,i}\rangle_{b,d} \right) + \\
 & \left(\sum_i c_{5,i} |\ell_{9,i}, \ell_{10,i}\rangle_{a,d} \right) \left(\sum_i c_{6,i} |\ell_{11,i}, \ell_{12,i}\rangle_{b,c} \right) \big) \quad (5.33)
 \end{aligned}$$

with and $\ell_{i,j} \in \mathbb{Z}$. Furthermore, one can use filters both in the pump and down-converted photon paths, such that $c_{i,j} \in \mathbb{C}$ with $|c_{i,j}| \leq 1$. The W-state (presented in figure 3 of the main text) is a special case of figure 5.15. A high-dimensional example is shown in figure 5.16b. It shows a setup for creating $n=3$ photon entangled state (with one photon acting as a trigger $|0_T\rangle$) with $d = 4$ terms in an asymmetric configuration ($|\psi\rangle = \frac{1}{2}|0_T\rangle(|0_a0_b0_c\rangle + |1_a0_b1_c\rangle + |2_a1_b0_c\rangle + |3_a1_b1_c\rangle)$). The state can be quantified by the Schmidt-Rank Vector (4,2,2) [149]. A variety of similar states can be written down in that form.

High-dimensionally entangled quantum states with more than 4 photons can also be created in this way, even though their structure is not in the form of a GHZ state, but surprisingly, calculating the full state requires Graph Theory and is described in [197].

Interestingly, the simplest special case of this technique presented here is a commonly used source of two-photon polarisation-entanglement. The so called *cross-crystal source* uses two crystals after each other, where the first one can create a horizontally polarized photon pair, the second one creates vertically polarized photon pairs [198]. Pumping both crystals at the same time and producing one pair of photons, one can create a $|\psi\rangle = \frac{1}{\sqrt{2}}(|H,H\rangle + |V,V\rangle)$ state. That technique can now be seen as a special case of a much broader technique to produce highly flexible high-dimensional multi-photon states in various degrees of freedom, by exploiting superposition of photon-pair origins and identifying of paths of photons.

While the examples are described in the photonic regime, the method of creating entan-

glement by path identity (or more generally, identity of *some* degree of freedom) can be generalised to other quantum entities. It can potentially be applied for generating entanglement in microwave superconducting cavities [188], atomic systems [199, 200], trapped ions [201] and superconducting circuits [202].

5.7 Discussion

Here I demonstrated how a computer can design new quantum experiments. The large number of discoveries reveals a way to investigate new families of complex entangled quantum systems in the laboratory. Several of these experimental designs have been successfully implemented in our labs [98, 158], others are being built at the moment. This line of research (in particular, experimental investigation of high-dimensional multipartite entanglement) would not be possible without the algorithm and the designs presented here. In contrast to human designers of experiments, MELVIN does not follow intuitive reasoning about the physical system, and therefore leads to the utilization of many unfamiliar and unconventional techniques that are challenging to understand intuitively. The algorithm can learn from experience (i.e. previous successful solutions), which leads to a significant speedup in discoveries of more complex experimental setups.

MELVIN can be applied to many other questions about the creation and manipulation of quantum systems, such as the search for more general high-dimensional transformations with different degrees-of-freedom and for different physical systems, or for efficient generation of other types of important quantum systems such as NOON-states [203]. In order to improve the efficiency of finding solutions, powerful techniques from artificial intelligence research can be applied, such as an evolutionary algorithm [204] (where the experiment and the resulting quantum state play the role of genotype and phenotype, respectively), reinforcement learning techniques [171, 205, 206] (by implementing a reward function depending on the closeness of the quantum state's properties to the desired properties) or entropy-based [207] and big-data methods [208, 209] (in order to find more unexpected solutions).

As I have shown in example 3, the algorithm can not only be used to find experimental designs with predefined properties, but it can also help to find general new ideas for experimental techniques and methods. It would be certainly interesting to find more examples such as entanglement by path identity.

6 Conclusion and Outlook

In this work I have summarized my efforts in investigate photons with complex spatial structures in extremal situations – such as large Hilbert spaces (high-dimensional entanglement), large real spaces (distribution over large distances) and small real spaces (focused beams with large topological charge).

In particular, I have performed an experiment using spatial mode of photons to create and confirm an 100-dimensionally entangled quantum state. This result shows the of spatial modes great potential as carrier of both classical as well as quantum information (chapter 3, and detailed summary in 3.4).

Afterwards, to understand whether this potential can be used in classical or quantum communication schemes, I performed three different experiments. From these experiments we conclude that information encoded in the spatial modes of light can be extracted – without adaptive optics, but with an adaptive image recognition algorithm – over 3 kilometers across Vienna and over 143 kilometers between two islands. Furthermore, quantum entanglement encoded in the spatial mode of photons can be detected after 3 kilometers of freespace propagation across Vienna, which confirms that single-photon spatial coherence and two-photon coherence is not significantly disturbed by the turbulent atmosphere (chapter 4, and detailed summary in 4.5).

In order to investigate high-dimensional and multipartite entangled states in the laboratory, I developed a computer algorithm which automatically designs quantum experiments. We were now able to generate and investigate complex quantum states in our laboratories, which before we only analysed on paper. The techniques used in computer-designed experiments are very unfamiliar. It lead us to the discovery of a novel, very general method for generating high-dimensional entanglement. This was – in my opinion – a successful example of machine-human synergy (chapter 5, and detailed summary in 5.7).

I would like to finish my thesis with a few questions and follow-up projects which I find genuinely exciting.

1) Small beams with large topological charge – and quantum entanglement

In chapter 2.3 I have shown an interesting non-paraxial effect that happens for small (focused) light beams with large OAM. There, the E_z component of the electric field cannot be neglected anymore, and significantly contributes to the intensity. In particular, for beams in superposition of $\pm\ell$, the perfect interference fringes vanishes as shown in figure 2.5.

6 Conclusion and Outlook

This interference visibility is a signature of entanglement of two-dimensional OAM modes and have been used to extract information about the entanglement in [44, 138] as well as by us in chapter 4.3. In particular, when the OAM values are very large OAM (as investigated in [44, 45]), the interference visibility would decrease due to the E_z component. That indicates that the entanglement is transformed into a more complex form, involving the z-direction of the electromagnetic field. It would be very interesting to understand that process in more detail.

2) Small beams with large topological charge – and matter waves

The paraxial wave equation (2.13) has led to paradoxical predictions for focussed light beams with large OAM, which we solved by applying techniques free from the paraxial assumption in chapter 2.3. Now the Schrödinger equation predicts the same paradoxical behavior for matter waves as the paraxial wave equation predicts for light. The question is what actually happens with matter waves described by the Schrödinger equation in this regime. Do photons and matter wave behave differently there, or are the results of the Schrödinger equation incorrect in that situation (similar as the paraxial wave equation's solutions were incorrect). It would be interesting to analyse this effect for electrons using the Dirac equation. If the same result occurs (vanishing of interference visibility), it necessarily is due to a different physical effect (such as a relativistic correction). It would be exciting if a similar phenomenon happens in other systems for entirely different physical reasons.

3) Maximal information encoded in an entangled system

In chapter 3 I show that two photons produced in parametric down-conversion are entangled in at least 100 dimensions, which is spread over 186 different spatial modes. While the dimensionality of entanglement is an interesting fundamental property of the quantum state, it does not necessarily correspond to the usefulness of the state in quantum protocols. Consider the state

$$|\psi_{(2+\epsilon)d}\rangle = \frac{|0,0\rangle + |1,1\rangle + \epsilon|2,2\rangle + \epsilon|3,3\rangle}{\sqrt{2+2\epsilon^2}}, \quad (6.1)$$

with small ϵ . Such a state is 4-dimensionally entangled, but the two photons barely share more than 1 bit of nonlocal information. The useful information is characterized by the ebits, or entangled bits, and is formally named entanglement-of-formation [210]. An intriguing question is how the non-local information encoded in photons can be increased and verified in a state-independent way, both experimentally and theoretically. Experimentally, a combination of polarization entanglement, spatial mode entanglement and frequency entanglement [211] might lead to very large numbers of ebits. But what is the experimental upper limit? A fascinating question is whether the information encoded at a single photon or the non-local information shared between two or more photons could be limited for some fundamental reason. From entropy considerations, Bekenstein has derived a fundamental upper bound of information in a given volume of space – the *Bekenstein bound* [212]. However, it is not clear whether this can be applied to our systems or whether other (stricter) bounds emerge due to other physical effects.

4) Fast, long-distance, free-space classical communication with spatial modes of light

In chapter 4.2 and chapter 4.4, we have shown that spatial modes of photons can indeed be transmitted over large distances through turbulent atmosphere (3 kilometers across Vienna, and 143 between two islands). The data rate of these experiments were very slow (in the order of bits per second). I believe that our results show that the modes survive the transport through the atmosphere, and can be used as a simple test to investigate the mode quality. Now very clever techniques need to be performed in order to extract the information. I would find it very exciting to see long distance, classical communication with data rates of Gigabits/sec or Terabits/sec, as it has been done in the laboratories [145]. A first experiment in this direction has already been reported which transmits 400Gbit/sec over 120 meters using OAM multiplexing [115], and clever adaptive optics techniques are being employed in simulated environments [142]. It would be exciting to see similar data rates for distances two or three orders of magnitude larger.

5) Potential of adaptive image recognition in classical and quantum communication

In our classical communication experiments, we have used an alternative method to extract the information from the light beam – in form of an adaptive image recognition algorithm (as explained in chapter 4.2.2). I would find it very interesting to understand better the potential of such algorithms both in classical and quantum communication. The great advantage is that they can adapt to turbulent atmosphere (for instance, using an artificial neural network as we have done), thus adaptive optics is not required. The drawback is that they cannot (as far as I can think of) work on a single photon level. I would find it very interesting to understand its potential, for instance, what is the minimum number of photons required to extract information, or how well can it adapt to strong turbulences. Maybe the most interesting question is whether there are situations when such adaptive algorithms (software-solution) can outperform adaptive optics systems (hardware solution). In a very recent article, some of these questions are being investigated for the first time [140]. There the authors investigate multiplexing of OAM modes in a simulated atmosphere, and the information is extracted via large, convolutional neural networks. They showed (for me surprisingly) very good initial results.

While such algorithms can not detect work on a single-photon level, they could still be useful for quantum experiments, for example when one uses single-photon sensitive cameras to image the spatial structures depending on the outcome of the entangled partner photon, as it has been done here [138], and similar as we have detected quantum entanglement after 3 kilometers in chapter 4.3. It would be very interesting to investigate adaptive image recognition algorithms in the quantum regime as well.

6) Experimental realisation of a scalable source for high-dimensional, maximally entanglement with spatial modes

In chapter 3, I have demonstrated the potential of spatial mode entanglement. Unfortunately however, the down-conversion process, which was used as the source of the high-dimensional entanglement, created higher-order modes with significantly lower probability. This leads to non-maximally entangled states. There is (to the best of my knowledge) no other source for photons which are high-dimensionally maximally entangled in their spatial modes.

6 Conclusion and Outlook

In chapter 5.6 I have explained a new technique to create entanglement (based on computer-designed experiments) – in particular a source for in principle maximally entangled high-dimensional states (see figure 5.14b). The source is – to the best of my knowledge – experimentally feasible, it is modular and importantly it’s scalable. I believe the experimental realisation of this design could enable very high-quality, high-countrate, tunable high-dimensional entangled states. It could be particularly interesting for long-distance distribution of high-dimensional entanglement. As the dimensionality of the entangled state is well-defined, one could use similar detection methods as we have shown in chapter 4.3 – which is not possible at the moment with down-conversion sources.

7) Automated search for new quantum experiments in other domains

In chapter 5, I have explained the computer algorithm MELVIN, which was essential for providing the designs of several experiments that have been performed recently in our laboratories. So far, I have applied it only on questions in photonic quantum optics (and there, mainly polarisation and OAM degree of freedom). I would find it very interesting to think about the implementation in other domains in quantum physics, such as superconducting cavities, trapped ions, NV centers. This could be useful in situations where there are experimental limitations (such as for the transformation of OAM modes of photons). Applying such algorithms to other domains has to be done in collaboration with experts in that domain, who understand precisely the experimental details and difficulties of their techniques.

8) Machine learning for finding new quantum experiments

The algorithm I presented in chapter 5 is based mainly on random search, with the possibility to extend its toolbox with previously useful experiments (explained in chapter 5.4.1). It would certainly be interesting to apply more powerful machine learning algorithm for similar tasks. In the best case, such an algorithm could find hidden structures in the experimental domains that are not (yet) known to the experimentalist. For that, I am recently working with the group of Hans Briegel from University of Innsbruck to apply a reinforcement learning algorithm [171] on the question of designing quantum experiments.

9) Surprising techniques inspired by computer-designed quantum experiments

In chapter 5.6 I have explained a technique for the creation of very general, high-dimensional, multi-particle entangled states. The idea was inspired by a solution by MELVIN which contained an *experimental trick* which I have never seen before and which we were able to understand and generalize. Obviously, it would be great find more such techniques.

In this example, I was analysing the experimental proposal of a very rare, unexpectedly high-dimensional entangled 3-photon state. Thus one idea would be to investigate other, very rare states and try to understand why they are so *elusive*. While feasible, this is very challenging because the computer-proposed experimental setups are counterintuitive thus quite difficult to understand.

Another idea to obtain surprising new techniques would be to modify the criteria when a setup is reported (as shown in figure 5.1). In the above examples, the task was to report

6 Conclusion and Outlook

for very concrete quantum states or quantum transformation. If one would instead select for certain properties of the states or transformations, the algorithm would have more freedom to surprise the human.

As a final, admittedly vague, thought: One necessary condition of a *surprising* fact is that it provides some new information. That has two consequences: 1) Surprisingness is subjective, 2) If one wants to create an algorithm which can find surprising facts or techniques more frequently, one probably needs to provide a large amount of information about that domain. And even if that amount of information is provided, it is likely very difficult to find a reasonable *surprisingness function*. There is exciting research on this topic, such as the definition and application of *Bayesian surprise* for specific domains [208, 213], or curiosity-driven exploration of the environment [214]. It would be amazing to find adequate methods which frequently lead to surprising results – and obviously this is not restricted to the domain of quantum experiments.

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Appendix

Experimental setups for other Cyclic Operations

3-cyclic OAM+Polarisation rotation

Experimental configuration:

$$\begin{aligned} \text{HWP}[\psi, a] &\rightarrow \text{Reflection}[XXX, a] \rightarrow \text{PBS}[XXX, a, c] \rightarrow \text{OAMHolo}[XXX, a, 2] \\ &\rightarrow \text{Reflection}[XXX, a] \rightarrow \text{PBS}[XXX, a, c] \rightarrow \text{BS}[XXX, a, b] \\ &\rightarrow \text{DP}[XXX, b, 2] \rightarrow \text{Reflection}[XXX, b] \rightarrow \text{BS}[XXX, a, b] \\ &\rightarrow \text{HWP}[XXX, b] \rightarrow \text{BS}[XXX, a, b] \rightarrow \text{DP}[XXX, b, 2] \\ &\rightarrow \text{Reflection}[XXX, b] \rightarrow \text{BS}[XXX, a, b] \end{aligned} \quad (7.1)$$

Operation:

$$|-2, V\rangle \rightarrow |-2, H\rangle \rightarrow |0, V\rangle \rightarrow |-2, V\rangle \quad (7.2)$$

The number in the ket stands for the OAM, H and V stand for horizontal and vertical polarisation.

6-cyclic OAM+Polarisation rotation

Experimental configuration:

$$\begin{aligned} \text{HWP}[\psi, a] &\rightarrow \text{Reflection}[XXX, a] \rightarrow \text{PBS}[XXX, a, c] \rightarrow \text{OAMHolo}[XXX, a, 2] \\ &\rightarrow \text{Reflection}[XXX, a] \rightarrow \text{PBS}[XXX, a, c] \rightarrow \text{BS}[XXX, a, b] \\ &\rightarrow \text{DP}[XXX, b, 2] \rightarrow \text{Reflection}[XXX, b] \rightarrow \text{BS}[XXX, a, b] \\ &\rightarrow \text{HWP}[XXX, b] \rightarrow \text{BS}[XXX, a, b] \rightarrow \text{DP}[XXX, b, 2] \\ &\rightarrow \text{Reflection}[XXX, b] \rightarrow \text{BS}[XXX, a, b] \end{aligned} \quad (7.3)$$

Operation:

$$|-4, H\rangle \rightarrow |-2, H\rangle \rightarrow |0, V\rangle \rightarrow |2, H\rangle \rightarrow |4, V\rangle \rightarrow |-4, H\rangle \quad (7.4)$$

The number in the ket stands for the OAM, H and V stand for horizontal and vertical polarisation.

8-cyclic OAM+Polarisation rotation

Experimental configuration:

$$\begin{aligned}
 \text{PBS}[\psi, a, b] &\rightarrow \text{BS}[XXX, b, c] \rightarrow \text{DP}[XXX, c, 1] \rightarrow \text{Reflection}[XXX, c] \\
 &\rightarrow \text{BS}[XXX, b, c] \rightarrow \text{Reflection}[XXX, b] \rightarrow \text{BS}[XXX, b, c] \\
 &\rightarrow \text{DP}[XXX, c, 1] \rightarrow \text{Reflection}[XXX, c] \rightarrow \text{BS}[XXX, b, c] \\
 &\rightarrow \text{OAMHolo}[XXX, b, 1] \rightarrow \text{PBS}[XXX, a, b] \rightarrow \text{HWP}[XXX, a] \quad (7.5)
 \end{aligned}$$

Operation:

$$|-1, V\rangle \rightarrow |-1, H\rangle \rightarrow |0, V\rangle \rightarrow \dots \rightarrow |2, H\rangle \rightarrow |-1, V\rangle \quad (7.6)$$

The number in the ket stands for the OAM, H and V stand for horizontal and vertical polarisation.

14-cyclic OAM+Polarisation+Path rotation

Experimental configuration:

$$\begin{aligned}
 \text{Reflection}[\psi, a] &\rightarrow \text{OAMHolo}[XXX, a, 2] \rightarrow \text{Reflection}[XXX, a] \rightarrow \text{OAMHolo}[XXX, a, -2] \\
 &\rightarrow \text{PBS}[XXX, a, b] \rightarrow \text{HWP}[XXX, a] \rightarrow \text{PBS}[XXX, a, b] \\
 &\rightarrow \text{Reflection}[XXX, b] \rightarrow \text{OAMHolo}[XXX, a, 2] \rightarrow \text{Reflection}[XXX, a] \\
 &\rightarrow \text{BS}[XXX, a, b] \rightarrow \text{DP}[XXX, b, 2] \rightarrow \text{Reflection}[XXX, b] \\
 &\rightarrow \text{BS}[XXX, a, b] \quad (7.7)
 \end{aligned}$$

Operation:

$$\begin{aligned}
 |0, H, a\rangle &\rightarrow |-2, H, b\rangle \rightarrow |-4, H, b\rangle \rightarrow |-8, H, b\rangle \rightarrow |10, V, b\rangle \\
 &\rightarrow |10, V, a\rangle \rightarrow |-6, H, a\rangle \rightarrow |8, H, a\rangle \rightarrow |6, H, b\rangle \\
 &\rightarrow |4, H, a\rangle \rightarrow |0, H, b\rangle \rightarrow |2, V, b\rangle \rightarrow |2, V, a\rangle \\
 &\rightarrow |2, H, a\rangle \rightarrow |0, H, a\rangle \quad (7.8)
 \end{aligned}$$

The number in the ket stands for the OAM, H and V stand for horizontal and vertical polarization, and a and b stand for the two different possible paths.

List of Publications

20. **Quantum Experiments and Graphs: Multiparty States as coherent superpositions of Perfect Matchings**
Mario Krenn, Xuemei Gu, Anton Zeilinger
arXiv:1705.06646 (2017).
19. **Orbital angular momentum of photons and the entanglement of Laguerre–Gaussian modes**
Mario Krenn, Mehul Malik, Manuel Erhard, Anton Zeilinger
Philosophical Transactions of the Royal Society of London A **375** (2087), 20150442 (2017).
18. **High-Dimensional Single-Photon Quantum Gates: Concepts and Experiments**
Amin Babazadeh, Manuel Erhard, Feiran Wang, Mehul Malik, Rahman Nouroozi, Mario Krenn, Anton Zeilinger
arXiv:1702.07299 (2017).
17. **Entanglement by Path Identity**
Mario Krenn, Armin Hochrainer, Mayukh Lahiri, Anton Zeilinger
Physical Review Letters **118** (8), 080401 (2017).
16. **Quantum communication with photons**
Mario Krenn, Mehul Malik, Thomas Scheidl, Rupert Ursin, Anton Zeilinger
Optics in Our Time (Springer International Publishing), 455–482 (2016).
15. **Twisted light transmission over 143 km**
Mario Krenn, Johannes Handsteiner, Matthias Fink, Robert Fickler, Rupert Ursin, Mehul Malik, Anton Zeilinger
Proceedings of the National Academy of Sciences **113** (48), 13648–13653 (2016).
14. **Quantum optical rotatory dispersion**
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13. **Cyclic transformation of orbital angular momentum modes**
Florian Schlederer, Mario Krenn, Robert Fickler, Mehul Malik, Anton Zeilinger
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12. **Automated Search for new Quantum Experiments**
Mario Krenn, Mehul Malik, Robert Fickler, Radek Lapkiewicz, Anton Zeilinger
Physical Review Letters **116** (9), 043019 (2016).

11. **On Small Beams with Large Topological Charge**
Mario Krenn, Nora Tischler, Anton Zeilinger
New Journal of Physics **18** (3), 033012 (2016).
10. **Multi-photon entanglement in high dimensions**
 Mehul Malik, Manuel Erhard, Marcus Huber, Mario Krenn, Robert Fickler, Anton Zeilinger
Nature Photonics **10** (4), 248 (2016).
9. **Physical meaning of the radial index of Laguerre-Gauss beams**
 William N Plick, Mario Krenn
Physical Review A **92** (6), 063841 (2015).
8. **Quantifying high dimensional entanglement with cameras and lenses**
 Paul Erker, Mario Krenn, Marcus Huber
arXiv:1512.05315 (2015).
7. **Twisted photon entanglement through turbulent air across Vienna**
Mario Krenn, Johannes Handsteiner, Matthias Fink, Robert Fickler, Anton Zeilinger
Proceedings of the National Academy of Sciences **112** (46), 14197-14201 (2015).
6. **Communication with spatially modulated light through turbulent air across Vienna**
Mario Krenn, Robert Fickler, Matthias Fink, Johannes Handsteiner, Mehul Malik, Thomas Scheidl, Rupert Ursin, Anton Zeilinger
New Journal of Physics **16** (11), 113028 (2014).
5. **Generation and confirmation of a (100x100)-dimensional entangled quantum system**
Mario Krenn, Marcus Huber, Robert Fickler, Radek Lapkiewicz, Sven Ramelow, Anton Zeilinger
Proceedings of the National Academy of Sciences **111** (17), 6243-6247 (2014).
4. **Real-time imaging of quantum entanglement**
 Robert Fickler, Mario Krenn, Radek Lapkiewicz, Sven Ramelow, Anton Zeilinger
Scientific Reports **3**, 1914 (2013).
3. **Quantum orbital angular momentum of elliptically symmetric light**
 William N Plick, Mario Krenn, Robert Fickler, Sven Ramelow, Anton Zeilinger
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2. **Entangled singularity patterns of photons in Ince-Gauss modes**
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1. **Quantum entanglement of high angular momenta**
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