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Abstract

Matter wave interferometry is a useful tool to demonstrate quantum mechanical properties for a broad class of matter [1–4]. With the newly constructed long-baseline universal matter-wave interferometer (LUMI) it is possible to carry out interference experiments with single atoms up to complex molecules and soon also heavy metal clusters [5,6]. The high sensitivity of the setup can further be used for advanced metrology on interfering particles [7–9]. We can sense inertial forces such as gravity or Coriolis [10] and measure molecular properties such as the magnetic susceptibility [11] or the static polarizability [12]. The principle of operation of this interferometer has already been established for molecules in previous near-field matter-wave interferometers [13,14]. However, LUMI is particularly well suited for high-mass interference and precision experiments due to the two-meter long baseline. This enables interference with molecules of an order of magnitude higher mass and a 100-fold increased force sensitivity compared to the predecessor. LUMI is therefore compatible with molecules up to 100.000 amu, while being able to sense forces as low as $F = 10^{-26}$ N. An electrode and a deflection magnet, which are installed in the experimental setup of the interferometer, can be used to deflect the interfering particles. In addition to the existing deflectors, the interferometer was expanded by a second magnetic system consisting of two coils in an anti-Helmholtz arrangement during the course of this thesis. The coils produce a tunable uniform gradient field and can be used to study atoms and molecules exhibiting a permanent magnetic moment. I will further describe the concept of matter wave interferometry in the context of LUMI and present recent results in the world of quantum macroscopicity: The superposition of the largest molecule (up to 28.000 amu) in a matter wave interferometer to date [15]. In a brief outlook I will discuss the planned updates of the interferometer. The new version of LUMI with three optical gratings will enable interference of metal clusters and thus push the current mass limit to even heavier molecules.

Zusammenfassung

Materiewelleninterferometrie ist ein nützliches Werkzeug um quantenmechanische Eigenschaften für eine breite Klasse von Materie nachzuweisen [1–4]. Mit dem neu konstruierten Long-baseline Universal Matter-wave Interferometer (LUMI) können Interferenzexperimente mit einzelnen Atomen bis hin zu komplexen Molekülen und bald auch schwere Metall-Cluster durchgeführt werden [5, 6]. Die hohe Empfindlichkeit des Interferometers kann ferner für Metrologie verwendet werden [7–9]. Einflüsse durch Inertialkräfte, wie die Gravitation oder die Corioliskraft [10], aber auch molekulare Eigenschaften, wie zum Beispiel die magnetische Suszeptibilität [11] oder die statische Polarisierbarkeit [12] können so zugänglich gemacht werden. Das Funktionsprinzip eines solchen Interferometers für Moleküle wurde bereits im vorangehenden Experiment demonstriert [13, 14]. LUMI eignet sich aufgrund der zwei Meter langen Grundlinie besonders gut für Interferenz- und Präzisionsexperimente mit hoher Masse. Dies ermöglicht die Interferenz mit Molekülen mit einer um eine Größenordnung höheren Masse und einer 100-fach erhöhten Empfindlichkeit gegenüber Kräften verglichen zum Vorgänger. LUMI ist daher mit Molekülen bis zu 100.000 amu kompatibel und kann Kräfte von nur $F = 10^{-26}$ N erfassen. Eine Elektrode und ein Ablenkungsmagnet, die im Versuchsaufbau des Interferometers installiert sind, können für metrologischen Anwendungen an den interferierenden Molekülen verwendet werden. Zusätzlich zu den vorhandenen Deflektoren wurde das Interferometer im Rahmen dieser Arbeit um ein zweites Magnetsystem erweitert, das aus zwei Spulen in einer Anti-Helmholtz-Anordnung besteht. Die Spulen erzeugen ein einheitliches Gradientenfeld und können verwendet werden um Atome und Moleküle mit einem permanenten magnetischen Moment zu untersuchen. Ich werde außerdem das Konzept der Materiewelleninterferometrie im Rahmen von LUMI beschreiben und aktuelle Ergebnisse zur Welt der Quantenmakroskopizität vorstellen: Die Quantenüberlagerung des bislang größten Moleküls (bis zu 28.000 amu) in einem Materiewelleninterferometer [15]. Weiters werde ich in einem kurzen Ausblick die geplanten Updates des Interferometers diskutieren. Die neue Version des Interferometers mit drei optischen Gittern soll zukünftig Interferenzexperimente mit Metallclustern ermöglichen und damit die aktuelle Massengrenze zu noch schwereren Moleküle verschieben.

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'Not only is the Universe stranger than we think, it is stranger than we can think.'

WERNER HEISENBERG

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Chapter 1

Introduction

1.1 The mystery of quantum mechanics

A central role in quantum theory is played by the wave functions as solutions of the Schrödinger equation and by the matter waves as proposed by Louis de Broglie [16]. Although the evolution of the wave-function is deterministic, quantum theory makes purely probabilistic statements for the observation of a system. Yet no other known theory can describe the world of atoms and molecules better than modern quantum physics. The far-reaching understanding of some of these quantum effects have opened many opportunities for technical applications such as quantum communication, quantum imaging, quantum sensing and quantum computing [17]. On the other hand there is the perception of reality with all the classical laws of physics as we can experience it every day. Our world seems to somehow differ from the quantum world, as we cannot observe delocalized cars or interference of footballs. Of course, one could argue that this reality, the world as we perceive it day by day, can be adequately explained by existing theories and yet the question of what happens in between these two regimes remains unanswered. What happens suddenly to all this *quantumness* when we take a look at our macroscopic world? What happens to all those things that are too big to act quantum and too small for our concept of macroscopicity? Is there a limit or a border and how is 'too

small' and 'too big' even defined? By mass? By size? Complexity or number of atoms?

These are just a few examples of the many questions that arise when thinking about the transition from quantum to classical reality and it is also these questions that have always concerned many physicists and philosophers since the beginning of quantum mechanics. Today it is still subject of modern research and despite all these efforts, there is still no clear, correct view of understanding quantum mechanics within the concept of the reality known to us [18–21]. The summarized unsolved questions arising at the border between the quantum and the classical word are known as the measurement problem. A famous example of the measurement problem is Schrödingers famous Gedankenexperiment about a cat in a box (fig. 1.1) [22].

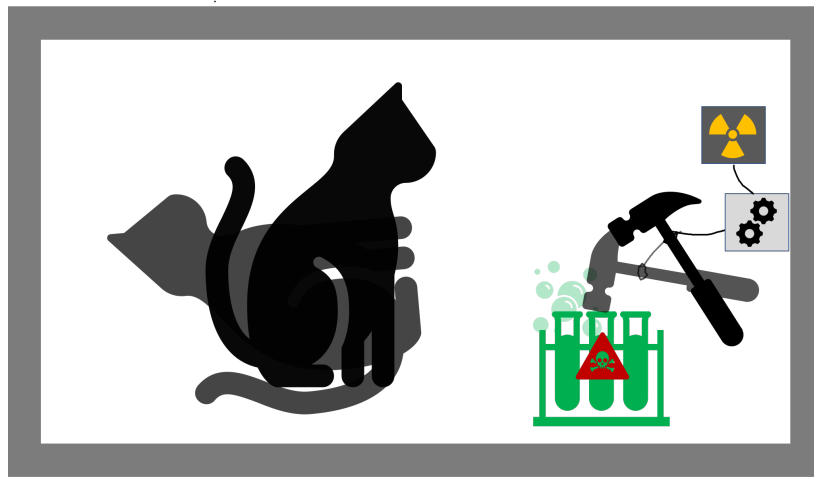


Figure 1.1: Schrödingers cat paradox: Schrödingers Gedankenexperiment places a living cat into a box together with a vial of a deadly poison. A hammer is mounted right above the vial and triggered by the decaying event of a radioactive atom. According to quantum mechanics, the atom is in a superposition state of decayed and not decayed. Since we do not know the state of atom exactly, we don't know if the vial is broken or not. Thus the cat is entangled with the quantum object and can, therefore, be described by a superposition state of dead and alive.

The paradox of Schrödinger's cat is about a cat sitting in an isolated box to-

gether with a mechanism that can kill the cat. For example a vial of poison and a hammer mounted on some releasing mechanics. The mechanics are then to be linked to a radioactive atom and released when the atom decays. According to quantum mechanics, the atom is in a superposition of decayed and un-decayed states. This entangles the macroscopic cat with the quantum state, being in superposition of a living cat and a dead cat. However, as soon as the box is opened, the observer can tell if the cat is dead or alive. The interpretations of the measurement problem mainly deal with the question of whether the collapse of the wavefunction was caused by the measurement or by something else or if there is any collapse at all. Schrödinger's cat is an exaggerated example for macroscopic quantum systems but it does well in demonstrating the arising problems. There are several different physical but also philosophical approaches to how the quantum theory with all its quirks could be interpreted. Philosophizing about such examples is one way to tackle the measurement problem but there is also the possibility to approach it experimentally. One way to do so is to start from where we know that the theory of quantum mechanics is valid and from there we gradually go to more and more macroscopic systems, until we reach a point where we cannot observe any quantum effects. Our approach to this is to use matter-wave interferometry and scale up the mass of the interfering particle. Due to the wave properties of matter we can create macroscopic quantum superposition in space in the interferometer. We use these cat-states to probe the validity of the quantum theory for a wide range of particles, ranging from isolated atoms to macroscopic molecules. The interferometric approach has the additional advantage that the highly sensitive experimental setup can be used for metrological experiments on the interfering particles. I will outline the underlying theory of matter-wave interferometry and quantum-enhanced metrology after a brief historical overview.

1.2 Matter-wave interferometry: Then and now

Starting from the very beginning The origins of quantum mechanics can be already found in the early attempts of Max Planck (1900) [23] with his idea of quantized energy. Later on, Max Planck's theory of blackbody radiation and quanta, an early version of quantum theory was successfully applied by Albert Einstein to explain the photoelectric effect and postulate the existence of photons (1905) [24], Stefan Pröppert (1913) [25] to calculate the magnetic moment of electrons. Furthermore, Niels Bohr (1913) [26] used Planck's constant to work out his model of the hydrogen atom and confirm Pröppert's value for the magnetic moment of electrons, nowadays known as the Bohr magneton. Some years later the theory of matter-waves was born when Louis de Broglie (1923) [16] extended the wave characteristics, exhibited by light, to any particle. Several physicists took part in shaping the modern theory of quantum mechanics. With Erwin Schrödinger (1926) [27] developing his wave-mechanical quantum theory, based on de Broglie's concept of matter-waves, Paul Dirac (1927) [28] deriving the relativistic wave equation and Max Born (1927) [29], Pascual Jordan (1927) [30] and Werner Heisenberg (1930) [31] working on a matrix-based theory of quantum mechanics. Since then, the non-relativistic Schrödinger equation has played a key figure in quantum mechanics [17]. The equation describes the evolution of a quantum-mechanical system. The linearity allows the existence of superposition states, leading to many apparent paradoxes [21]. The arising discrepancies between quantum and classical reality have been an inspiration for several thought experiments [22, 32, 33].

The beginnings of matter-wave interferometry The first matter-wave experiments were conducted by Davisson and Germer in 1927, who demonstrated the wave nature of electrons by diffraction [34]. The experiment was first suggested by Walter M. Elsasser (1925) [35] and initially meant to study the surface of nickel rather than verifying de Broglie's hypothesis [36]. In the same year,

Thomson and Reid independently of Davisson and Germer made the same discoveries through diffracting electrons from a crystal [37], for which Davisson and Thomson shared the Nobel prize in 1937 [38].

Another famous experiment is Young's double-slit with light [39]. It is historically first demonstration of the wave nature of light and serves today for massive particles, too. Double slit experiments were realized for electrons [40,41], neutrons [42,43], atoms [44,45] and molecules [46], confirming again the theory of matter-waves. The earliest example of molecular matter-wave interference experiment can be traced back to 1930 and Estermann and Stern, diffracting H_2 molecules at a crystal surface [47]. The first concept for an atom interferometer was then published in a patent by Altshuler and Franz (1973) [48], subsequent proposals followed different groups [49–54]. The diffraction of sodium atoms at a nano-fabricated grating at the M.I.T. (1988) [55] can be attributed to as the first atom interference at a nanomachined grating. A few years later the principle of matter-wave interference was extended to the He dimer and trimer [56].

The first step towards macromolecular matter-wave interferometry was done by Arndt et al. in 1999 with the diffraction of C_{60} [57] and later also C_{70} [58]. After the effect of the grating-particle interaction was demonstrated [59,60], diffraction from a light grating started became more important in the field of molecular matter-wave interferometry. The idea of using a standing light wave as a diffraction mechanism for electrons was already proposed by Kapitza and Dirac in 1933 [61] in the context of Bragg diffraction. Since then it has been applied to electrons [62], atoms [63–69] and molecules [13,70,71], with the light-matter interaction being different for all these species. While electrons passing a standing light wave can be described by the ponderomotive potential [72], diffraction of atoms and molecules depends on the phase-shifts due to the interaction of the induced electric dipole moment and the periodic electric field [73,74].

A major step towards high-mass interference came with near-field interferometry and the Talbot effect. The lens-less self-imaging effect for diffraction of light at a grating was first discovered by Talbot in 1832 [75] and soon established in the optical domain [76, 77]. Sharing the same motivation, the Talbot effect also holds for matter-wave diffraction experiments [1, 78–83]. The mass dependence of the grating periods for near-field interferometers, which is proportional to $1/\sqrt{m}$, is particularly advantageous for high-mass matter-wave interferometry, compared to the $1/m$ dependence of far-field interferometry.

The incoherent molecular sources made it furthermore necessary to also make use of Lau’s discoveries from 1948 [84] of lens-less self imaging with incoherent illumination. The Lau effect, originating in the field of light optics [85–87], is the extension of the aforementioned Talbot effect. The first Talbot-Lau interferometers (TLI) in the near-field for atoms [88–90] and molecules [1, 70, 81] followed.

The combination of the near-field Talbot effect for incoherent sources and diffraction at optical gratings led to the first implementation of a Kapitza-Dirac-Talbot-Lau interferometer (KDTLI) in Vienna [13]. The KDTLI was able to show quantum superposition for macro-molecules with masses up to 10.000 amu [4, 14, 91–93]. An extension of the KDTLI experiment is the long-baseline universal matter-wave interferometer (LUMI) [6], with which we were able to observe superposition of molecules with masses up to 28.000 amu and consisting of up to 2000 atoms [15].

Chapter 2

Theory

The experiments in this thesis build on quantum optics and matter-waves physics. Since this topic has already been summarized in more detail [5, 14, 92, 94, 95].

2.1 Matter-wave physics

As proposed by Louis de Broglie in 1923 [16], we can assign a wavelength to any massive particle in motion, the so-called de Broglie wavelength $\lambda_{dB} = h/mv$, where h is Planck's constant, m the mass and v the center-of-mass velocity of the particle. This has enabled applications such as electron imaging or neutron optics but also fundamental tests of quantum mechanics or relativity theory. A possible tool to demonstrate wave-related quantum behavior is to demonstrate matter-wave interference. The key idea is analogous to Young's double slit experiment. When the matter-wave travels along two or more indistinguishable paths, it will pick up phase differences and interfere either constructively or destructively when the wavelets are recombined. According to Born's rule, the probability density of finding a particle at time t at the position (x, y, z) is given by the square of the wave function $|\Psi(x, y, z, t)|^2$ [96]. Therefore, the accumulated phases translate into a modulation of the particle density. Given

the time-independent Schrödinger equation, we can rewrite the equation for matter-waves such that it has the same form as the Helmholtz equation:

$$(\Delta + k_{dB}^2)\Psi(\vec{x}) = 0, \quad (2.1.1)$$

with $k_{dB} = 2\pi/\lambda_{dB}$. We can treat diffraction and interference phenomena of matter-waves analogously to wave optics according to Huygens' principle [97], while keeping in mind dispersive effects due to velocity-dependent de Broglie wavelengths.

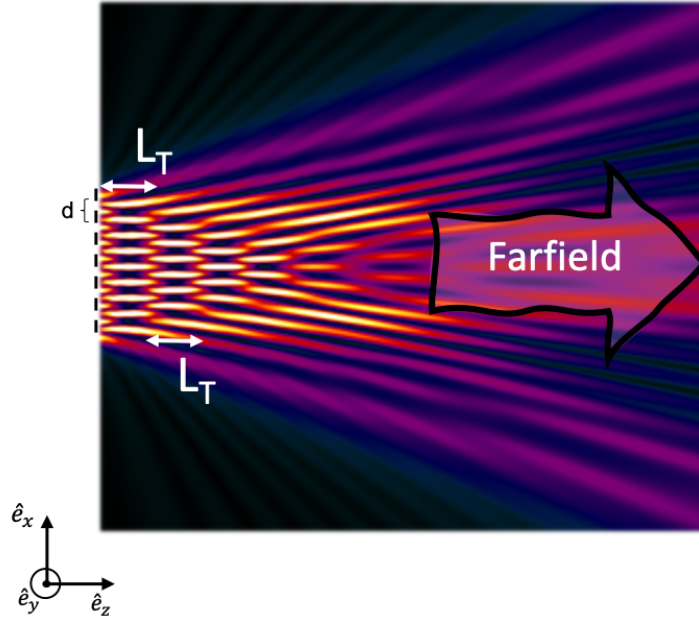


Figure 2.1: Simulation of the intensity distribution after diffraction of a plane wave at a single diffraction grating where you can see the transition from the near-field to far-field regime. While the curvature of the wave fronts plays a role in the near-field Fresnel diffraction, they can be approximated by plane waves after a certain distance $\sim a/\lambda_{dB}$, leading to the Fraunhofer diffraction in the far-field. The near field pattern is dominated by the periodic self-images of the grating at intervals of the Talbot length.

Near-field vs. far-field For the diffraction of a plane wave at a grating, we can distinguish between near-field and far-field effects. The Long Baseline Universal

Matter-wave Interferometer (LUMI), which is the subject of this thesis, operates in the near-field regime. The important difference is that when solving the Kirchhoff-Fresnel integral for the propagation of waves behind a grating, in the near-field the curvature of the wavelets has to be taken into account. This leads to the formation of a Talbot carpet consisting of self-images of the diffraction grating. The approximated transition from the near-field to the far-field takes place at $\sim a/\lambda_{dB}$, depending on the aperture size a and on the de Broglie wavelength as illustrated in fig. 2.1.

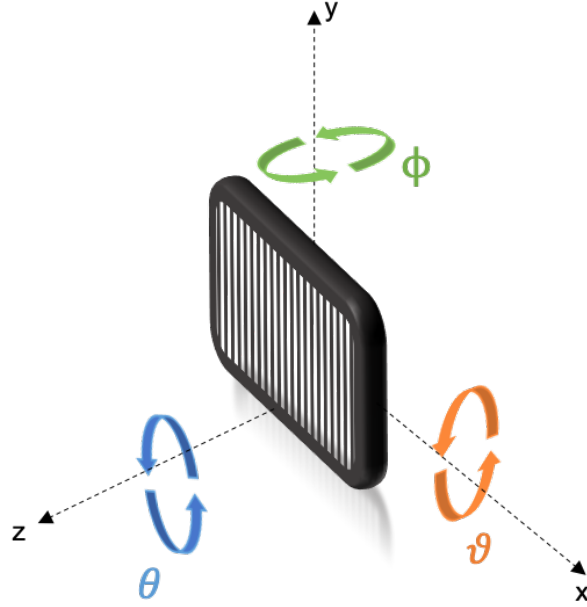


Figure 2.2: Definition of the rotational degrees of freedom for the gratings. A rotation around the molecular axis z is defined by a roll θ (blue), around the vertical axis by a yaw ϕ (green) and around the transversal axis x by a pitch ϑ (orange).

The near-field approach has significant advantages for matter-wave interferometry compared to the far-field, especially when it comes to massive molecules. Due to the Talbot length, the required grating periods for massive molecules scale with $1/\sqrt{m}$ compared to $1/m$ dependence of far-field diffraction angles,

for a given interferometer length. This means that for a fixed setup length and grating period the near-field interferometer can cope with smaller de Broglie wavelengths and is, therefore, better suited for massive molecules. Another important difference comes with the coherence requirements, which are more relaxed for the Talbot-Lau interferometer, allowing to work with spatially incoherent sources. However, the additional gratings come with strict alignment requirements. In addition to having equal longitudinal distance, the three gratings must have the same period and must be aligned translationally and additionally in terms of the three rotational axes yaw ϕ , pitch ϑ and roll θ (fig. 2.2) with respect to each other. The longitudinal separation for N illuminated slits is approximated as $\Delta L < L/N$ [5]. The alignment requirements in LUMI for a particle beam of $1 \times 1 \text{ mm}^2$, a grating separation of $L = 0.98 \text{ m}$ and grating period of $d = 266 \text{ nm}$ can be given as follows: $\Delta L = 260 \text{ } \mu\text{m}$, $\Delta d \leq 0.4 \text{ } \text{\AA}$, $\Delta\vartheta \leq 520 \text{ mrad}$, $\Delta\phi \leq 16 \text{ mrad}$ and $\Delta\theta \leq 0.08 \text{ mrad}$ [5, 14, 92, 98].

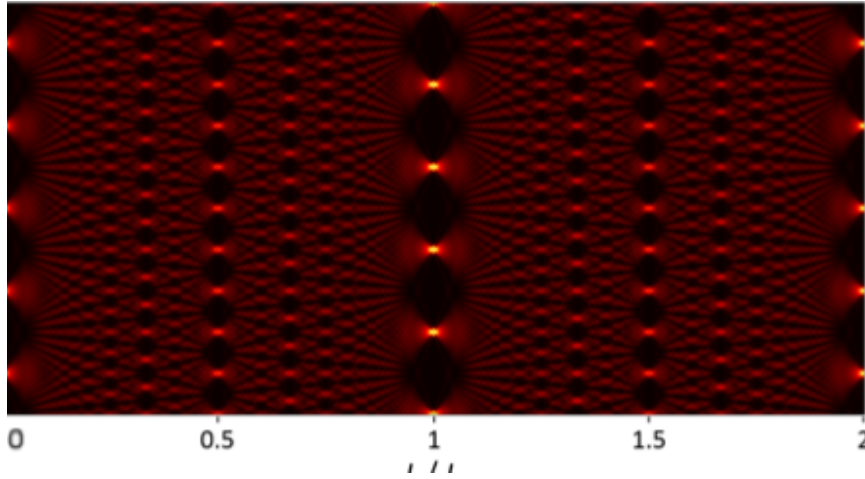


Figure 2.3: Detailed simulation of Talbot-carpet behind a single grating. Self-images of the diffraction grating reoccur periodically at integers distances of the Talbot length $L_T = d^2\lambda$. While even multiples are identical with the original structure, odd multiples appear shifted by half a period. Fractional revivals with periods of $1/p$ can be observed for fractional distances q/pL_T , with $q, p \in \mathbb{N}$.

Coherence requirements Observing interference of waves generally requires some coherence. Fast massive particles in our interferometers have a very short de Broglie wavelength in the order of a few femtometers. Accordingly, in order to observe interference with matter, very small grating periods and narrow collimated molecular beams are required. In terms of matter-wave interferometry, the coherence requirements are primarily placed on the molecular beam source. While each point of an extended source can create individual interference patterns, they come without phase-relation and thus wash out the contrast. The spatial coherence X_c defines minimum geometrical requirements to be able to observe interference fringes. It must be sufficiently large to cover a significant extent of the diffracting element, for example the grating period d . According to the van Cittert–Zernike theorem, the spatial coherence of an incoherent source has the same form as the diffraction pattern behind the source aperture a when illuminated with plane waves [99]. The spatial coherence at a distance L behind an incoherent source with an aperture size a is given by

$$X_c = \frac{2L\lambda_{dB}}{a} > d. \quad (2.1.2)$$

This means that small de Broglie wavelengths require either tiny source apertures or large flight distances to fulfill this condition. large. The second coherence condition that must be fulfilled regards the longitudinal coherence or also temporal coherence L_c , which is given by the Fourier transform of the spectral density, according to the Wiener-Khinchin theorem [99]. It indicates how similar the frequencies are. The spread in wavelengths, i.e. in velocities, must but sufficient narrow to avoid washing out of the interference fringes. It is therefore directly connected to the velocity spread $v/\Delta v$ of the molecular source and for a Lorentzian velocity distribution given by

$$L_c = \frac{\lambda_{dB}^2}{\Delta\lambda_{dB}} = \lambda_{dB} \frac{v}{\Delta v} > n\lambda_{dB}. \quad (2.1.3)$$

The ability to observe the n^{th} diffraction order of the far-field interference

pattern requires the coherence length to be greater than $n\lambda_{dB}$. In comparison to the spatial coherence the longitudinal coherence cannot be increased by changing the source geometry. To obtain a better temporal coherence the velocity spread of the particle beam must be narrowed or postselected. Velocity classes within the particle beam can for instance be gravitationally selected by using delimiting horizontal slits or time-of-flight measurements [74].

Far-field diffraction It was shown grating diffraction in the far-field for massive molecules like C_{60} [57] and C_{70} [58] with de Broglie wavelengths on the order of a few picometers. Since fast massive molecules can come with even smaller wavelengths, the above coherence requirements demand impracticable conditions for far-field diffraction with sub-microradian collimation and diffraction angles. This would limit the beam flux, which must be compensated by improved detector schemes to keep the signal-to-noise ratio high. Slowing the particles can help to path the way for more massive molecules. Reducing the forward velocity, however, increases the divergence angle which again must be compensated by a narrow collimation. A more useful approach is to cool the particles, reducing the motion in all three dimensions and also the internal excitations.

Talbot effect The lens-less self imaging was first observed by William Henry Fox Talbot in 1836 [75] and is well known as the Talbot effect in wave optics [100]. It describes the recurring self-imaging of an illuminated periodic structure at integer n distances behind this structure, the so-called Talbot length $L_T = 2d^2/\lambda$, with d the period of the structure and $\lambda = \lambda_{dB}$ the wavelength or the de Broglie wavelength in case of matter-waves. At half integers the same periodic structures can be observed, while being shifted with half a grating period compared to the original structure (fig. 2.3). Hence, the Talbot length can also be defined as

$$L_T = \frac{d^2}{\lambda}, \quad (2.1.4)$$

which will be used as the definition of choice throughout this thesis. Additionally, smaller periods can be observed at fractional distances $n = q/p$, with $q, p \in \mathbb{N}$, in the fractional Talbot effect [101]. However, the smallest fractional period is limited by the smallest feature of the diffraction grating. The coherent near-field self-imaging can be observed whenever a periodic structure is illuminated by a monochromatic plane wave. In principle, we can have Talbot interference by using a single grating.

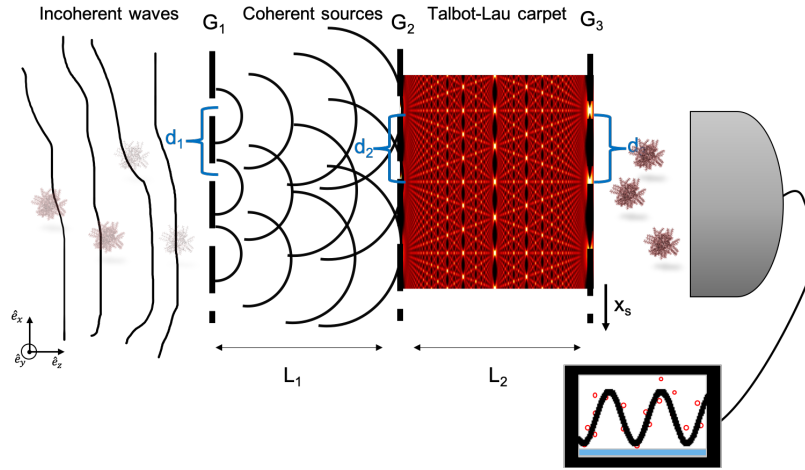


Figure 2.4: A schematic illustration of a TLI interferometer with three gratings. The matter-waves are initially arriving incoherent at the first grating. Due to the confinement of the waves within the slits, each of the slits acts as a coherent source for the second grating. When the waves arrive at the second grating, they are delocalized over at least two neighboring slits. If the distances $L_{1,2}$ and the grating periods $d_{1,2,3}$ fulfill the resonance conditions (eq.2.1.5 - 2.1.7), coherent diffraction at the second grating leads to the self-imaging effect described by Lau. The third grating can be scanned transversely (Δx_s) to reveal the periodic intensity distribution.

Lau effect The challenge for matter-waves is that molecular sources are in general polychromatic, generating a mixture of plane waves facing random directions. So, most of these sources come with a poor longitudinal and transverse coherence. Even though the Talbot effect occurs for every single molecule, the addition of all those slightly different shifted self-images will wash out the overall contrast. There is a way to get around this by using a second grating in

front of the diffraction grating, taking advantage of the Lau effect.

In 1948 Ernst Lau described the coherent self-imaging effect for incoherent illumination by using two gratings instead of one [84]. He found that one can obtain high contrast self-imaging pattern if the distance z_0 between the two gratings equals to $nd^2\lambda = L_T$ [86], which can indeed be explained purely geometrically: The waves from the incoherent source arrive at the slits of the first grating without any phase-relation. The slits of the first grating provide sufficient spatial confinement to prepare coherence when the waves arrive at the second grating. However, these waves are incoherent with respect to each other and their intensities have to be added up. At integer distances of nd^2/λ the maximum of these interference fringes coincides exactly at the position of the slits of the second grating such that the secondary waves that are generated in this slits share the same phase. The Lau effect extends the self-imaging Talbot effect for incoherent illumination¹. As with the Talbot effect, the Lau effect was initially observed for light but the same physics applies to matter-waves [14].

Talbot-Lau Interferometer LUMI is not a two-grating but a three-grating interferometer. While the purpose of the first two gratings is for the Talbot-Lau diffraction, the third grating is used as a scanning mask to reveal the periodic pattern. This grating could in principle be replaced by a surface on which the molecular density pattern is collected and imaged [102]. Also, there is no imperative that the gratings periods or the distances between the gratings are equal. However, a proper calculation of the Talbot-Lau effect reveals that two resonance conditions for the grating periods $d_{1,2,3}$ and the distances $L_{1,2}$ (fig. 2.7) have to be met, in order to obtain high-contrast patterns [103]

$$\frac{L_1 + L_2}{L_2} \frac{d_2}{d_1} = r_1, \quad (2.1.5)$$

¹Strictly speaking, the Talbot effect is a special case of the Lau effect where the distance between the source and the grating approaches infinity.

$$\frac{L_1 + L_2}{L_1} \frac{d_2}{d_3} = r_2, \quad (2.1.6)$$

with $r_{1,2} \in \mathbb{N}$. This means that for fixed grating periods $d_{1,2}$ the period of the third grating is restricted to

$$\frac{1}{r_1} \frac{d_2}{d_1} + \frac{1}{r_2} \frac{d_2}{d_3} = 1. \quad (2.1.7)$$

LUMI is operated in the 1:1:1 scheme, where $L_1 = L_2 = L$ as well as $d_1 = d_2 = d_3 = d$, hence $r_1 = r_2 = 2$. Even though all grating periods are equal, the symmetric arrangement relies on the fractional Talbot effect rather than on the integral Talbot effect. However, since the magnification factor $L_1 + L_2/L_2 = 2$ and the fractional order 2 cancel out, we can observe the integral pattern with the period d at integer distances of the Talbot length.

Diffraction gratings We differentiate between nano-fabricated gratings and optical gratings and between their type of interaction with the particles, which can either be a phase modulation or an amplitude modulation. A nano-fabricated material grating is usually referred to as a transmission mask and thus falls always into the category of an absorptive grating. It can be described as a pure transmission grating if the Van-der-Waals interactions between the particles and the grating walls can be ignored. The effect of the Casimir-Polder potential can lead to a position-dependent phase shift with up to hundreds of the grating period for highly polarizable molecules near the grating walls. The significance of the dispersive shift depends on the polarizability but also on the velocity of the beam. The slower the particles and the longer the interaction time, the higher the range of possible shifts and therefore the loss of contrast due to phase-averaging.

Optical gratings Optical phase gratings are useful matter-wave beam splitters for slow beams of polarizable molecules. The use of a standing light wave

was first described by Kapitza and Dirac in 1933 [61] for Bragg diffraction of electrons with the ponderomotive force explaining the interaction between the electrons and the standing light wave [72]. For atoms and molecules it is the interaction between the optical field and the polarizability α_ω [73] that causes the periodic shifts. The narrow line-widths of atoms with absorption cross-sections in the order $\sigma_{abs} = 10^{-9} \text{ cm}^2$ allow to change internal states by using near-resonant light. In contrast to that, the line-widths of molecules can extend over nanometers, resulting in absorption cross-sections in the order of $\sigma_{abs} = 10^{-15} \text{ cm}^2$. However, the off-resonant standing light wave induces an oscillating electric dipole moment which again acts with the periodic laser field [4]. Assuming that the standing light wave is formed by a retro-reflected laser along the x-axis with a TEM_{00} mode, the periodic potential of the light field that is seen by the molecules flying along the z-axis and in the vertical center ($y = 0$) is given by

$$V(y, z) = -\frac{2\pi\alpha_\omega}{c} I(y, z), \quad (2.1.8)$$

with the time-averaged intensity

$$I(y, z) = \frac{8P}{\pi w_y w_z} e^{(-\frac{2y^2}{w_y^2} - \frac{2z^2}{w_z^2})} \sin^2 \pi \frac{x}{d}, \quad (2.1.9)$$

where $d = \lambda_L/2$ is the grating period, $w_{y,z}$ are the beam waists, P is the laser power and z is the flight direction of the molecules.

The electric field induces an oscillating dipole moment in the molecule, which in turn interacts with the light field via the conservative optical dipole force. The position-dependent shift accumulated by molecules with a centre-of-mass velocity v_z can be calculated by integrating the field along a straight line

$$\Phi(x) = -\frac{1}{\hbar} \int_{-\infty}^{\infty} V(x, v_z t) dt = \phi_0 \sin^2 \pi \frac{x}{d}, \quad (2.1.10)$$

with a maximum shift Φ_0 that can be acquired

$$\Phi_0 = \frac{8}{\sqrt{2\pi}} \frac{\alpha_\omega}{\hbar c} \frac{P}{w_y v_z}. \quad (2.1.11)$$

Whether the optical grating acts amplitude or phase-modulating in depends on the optical polarizability α_ω , absorption cross section σ_{abs} and ionization energy E_i of the particles as well as on the intensity I and photon energy $\hbar\nu$ of the standing light wave [14]. The number of photons that can be absorbed in the anti-nodes of the optical grating is given by

$$n_x = \frac{\sigma_{abs}}{\hbar\nu} \int_{-\infty}^{\infty} I(x, v_z t) dt = n_0 \sin^2 \pi \frac{x}{d}, \quad (2.1.12)$$

with a maximum number of possible absorbed photons n_0 in the anti-nodes:

$$n_0 = \frac{8}{\sqrt{2\pi}} \frac{\alpha_\omega \lambda_L}{\hbar c} \frac{P}{w_y v_z}. \quad (2.1.13)$$

Each absorbed photon leads to a momentum transfer of $\hbar k$. If the molecule stays within the beam and re-emission of a photon does not occur on the time scale of the interferometer, the absorption of a photon can be treated as a stochastic phase shifts. If the absorption of a single photon is enough to ionize or fragment the molecule, the standing light wave can be used as a transmissive grating. In both schemes of the optical gratings, phase or amplitude modulation, the shifts are position-dependent and rely on the periodic pattern generated by the standing light wave. This in turn leads to the periodic self-imaging effect, similar to those from the material grating. [14]

KDTLI vs. TLI We distinguish between Talbot-Lau interferometry (TLI) consisting of three absorptive gratings and Kapitza-Dirac-Talbot-Lau interferometry (KDTLI) where we replace the second grating with an off-resonant optical phase grating (fig. 2.5). Both schemes have their justified application in matter-wave interferometry. In the first place the optical grating eliminates the highly

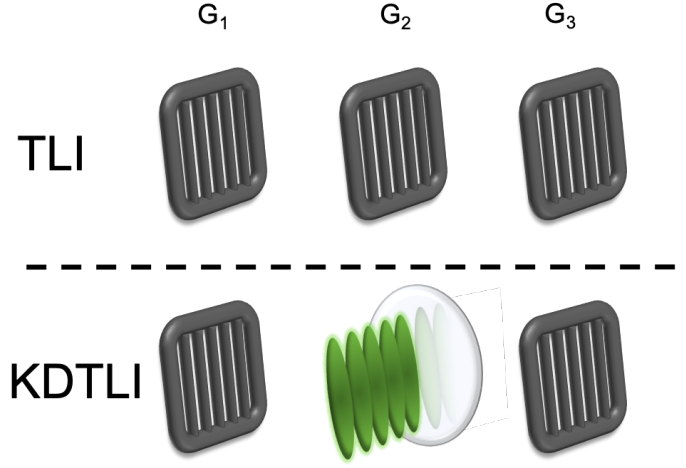


Figure 2.5: Schematic illustration of the difference between a Talbot-Lau interferometer (TLI) with three nano-fabricated material gratings and a Kapitza-Dirac-Talbot-Lau interferometer (KDTLI) with two material gratings and the second grating replaced with a standing light wave. While the optical phase-grating works for polarizable and slow molecular beams, the TLI scheme is limited to less polarizable molecules, due to the grating-molecule interactions.

dispersive Van-der-Waals interaction between the grating and the molecules. The plots depicted in fig. 2.6 show the expected visibility of a TLI/KDTLI scheme and the effects of the material-grating interaction for two different polarizable molecules. The optical grating also increases the transmission of the particles without any risk of clogging. However, it can only be used for highly polarizable molecules with sufficiently low velocities, which is often true for large molecules with polarizabilities in the order of $\text{\AA}^3$. In contrast to that, material gratings provide universality while being better suited for fast beams and less polarizable atoms or small molecules, since the Casimir-Polder effect is much weaker due to their low polarizabilities and the short interaction times. For the same reason very high laser intensities would be required to achieve sufficient shifts with optical phase gratings, which is the dominant exclusion criterion.

Since the waves at the first and the third grating come without phase-relation, the additional shifts due to the Casimir-Polder potential play no further role for the interference. They have to be absorptive to prepare coherence (G_1) and to

probe the interference pattern with a transmission mask (G_3) However, there are reasons to use three optical gratings worthwhile. First of all, if we want to take even larger molecules, we will need a smaller grating period at some point, assuming that the length of the interferometer stays the same. It is very difficult to get three identical and so precisely nano-fabricated gratings that meet all the requirements for the interferometer. The smaller grating periods also come with a higher risk of clogging. Optical gratings are realized by retro-reflecting laser beams and it is easy to generate three identical gratings using a single laser system. Already available laser systems providing high photon energies with sufficient power make optical depletion gratings suitable to act as absorptive gratings for metal clusters. A more detailed description can be found in the outlook of this thesis, where an all-optical setup is discussed as a part of the planned upgrade for LUMI. [104]

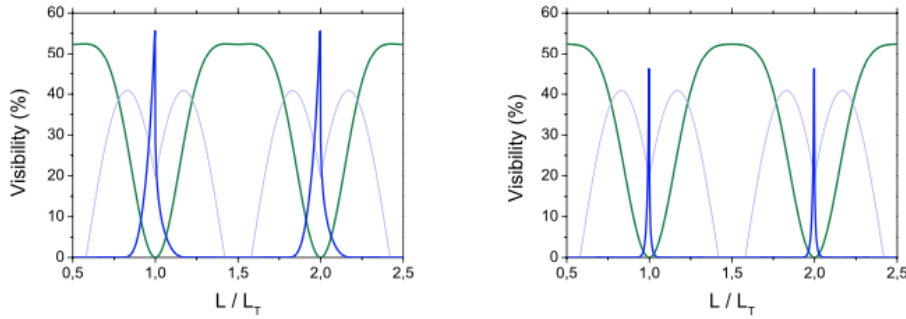


Figure 2.6: Comparison between the expected visibility in a TLI and KDTLI setup for C_{60} (left) and perfluoralkylated nanospheres (right). The light blue line, where the gratings are assumed to be perfect binary transmission gratings and the blue line, including the Casimir-Polder interaction, correspond to the TLI setup with three material gratings and the green line to the KDTLI setup with $G_{1,3}$ as material gratings and G_2 as an optical phase grating. The dispersive phase shift of the grating-molecule interaction narrows the domain of the high contrast, most for the molecule from the right plot, which has a higher polarizability than C_{60} . This can be circumvented when using an optical grating as the G_2 as can be seen in both cases. Image taken from reference [104].

2.1.1 A phase-space description of a (Kapitza-Dirac-)Talbot-Lau interferometer

A proper modeling of the molecular density pattern at the location of the third grating is required to link our experimental results to the quantum wave property of the molecules. A phase-space description using the Wigner-function is useful for this purpose. A complete calculation describing the transverse motion of a particle in the TLI/KDTLI has already been carried out in several publications [14, 95, 98, 104, 105] of which I will summarize only the key results. Since the Wigner-function is the quantum version of the classical phase-space density function, it can be used to directly compare quantum and classical models.

Wigner function The quantum state can be expressed as a density matrix, which is at the heart of the Wigner function in

$$w(x, p) = \frac{1}{2\pi\hbar} \int ds e^{ips\hbar} \langle x - \frac{s}{2} | \rho | x + \frac{s}{2} \rangle. \quad (2.1.14)$$

Here p and x are the momentum and position of the particle. We have limited the quantum description to the transverse motion x . This formulation allows a straightforward calculation of the transverse density pattern through the entire interferometer. The free evolution during time is a shear in phase space, given by

$$w_t = w_0 \left(t - \frac{t}{m} p, p \right), \quad (2.1.15)$$

with w_0 the initial Wigner-function and m the particles mass.

Grating transformation The effect of the gratings onto the quantum state can be calculated via the transformation

$$w'(x, p) = \int \int dx_0 dp_0 K(x, p; x_0, p_0) w_0(x_0, p_0), \quad (2.1.16)$$

where the kernel $K(x, p; x_0, p_0)$ relates to the grating transmission function $t(x)$ by [14]

$$K(x, p; x_0, p_0) = \delta(x - x_0) \frac{1}{2\pi\hbar} \int ds e^{i(p-p_0)s/\hbar} t\left(x - \frac{s}{2}\right) t^*\left(x + \frac{s}{2}\right). \quad (2.1.17)$$

The periodic grating allows for Fourier decomposition and the spatial density distribution after passing through the entire interferometer can be expressed as a function of the transversal shift of the third grating x_s

$$S(x_s) = \sum_m A_m^* B_{2m} \left(m \frac{L}{L_T}\right) C_m e^{2\pi i m x/d}, \quad (2.1.18)$$

where A_m^* , B_{2m} and C_m are the Fourier coefficients of the first, second and third grating. The first and the third grating are material gratings but the phase they imprint does not enter the form of the final interference pattern. The transmission can therefore be treated by a binary function. The rectangular shape of the transmission function then leads to the following form the Fourier-coefficient [14]

$$A_m = C_m = f \operatorname{sinc}(\pi m f), \quad (2.1.19)$$

where f is the opening fraction of the grating.

Material Talbot coefficient The Fourier-coefficient of the second grating B_{2m} , also known as the Talbot-coefficient can be written as

$$B_l(\xi) = \sum_j b_j b_{j-l}^* e^{i\pi\xi(l-2j)}. \quad (2.1.20)$$

Assuming a retarded Casimir-Polder potential $V(r) = -C_4/r^4$ the summands b_j can be calculated as [14]

$$b_j = \frac{1}{d} \int_{-s'/2}^{s'/2} dx \exp \left[-\frac{ib}{\hbar\nu} \left(-\frac{C_4}{r_-^4} - \frac{C_4}{r_+^4} \right) \right], \quad (2.1.21)$$

while b represents the grating depth and $r_{\pm} = fd/2 \pm x$.

Optical Talbot coefficient The modular design of LUMI allows us to replace the second grating with an optical phase grating in situ. The corresponding Talbot coefficient for the KDTLI scheme can be calculated considering the coherent effect of optical dipole force as well as the absorption events, which can be expressed by Bessel functions J_K [14]

$$B_k(\xi) = e^{-\beta_2(\xi)} \left(\frac{\beta_1(\xi) + \beta_2(\xi)}{\beta_1(\xi) - \beta_2(\xi)} \right)^{k/2} J_k \left[-\operatorname{sgn}(\beta_1(\xi) - \beta_2(\xi)) \sqrt{\beta_1^2(\xi) - \beta_2^2(\xi)} \right]. \quad (2.1.22)$$

Here, the coherent phase modulation $\beta_1(\xi)$ and a superposition from momentum kicks $\beta_2(\xi)$ of both sides of the standing light wave due to the photon absorption are given by

$$\beta_1(\xi) = \phi_0 \sin \left(\frac{\pi\xi}{d} \right) \quad (2.1.23)$$

and

$$\beta_2(\xi) = n_0 \sin^2 \left(\frac{\pi\xi}{d} \right). \quad (2.1.24)$$

Using these coefficients we can calculate the sinusoidal fringe visibility as the first order of the Fourier signal can be compared with the experiments.

$$V = 2 \operatorname{sinc}^2 \pi f B_2 \left(\frac{L}{L_T} \right). \quad (2.1.25)$$

Since the fringe pattern can be approximated very good with a sinusoidal function, we can also approximate the visibility with [14]

$$V \approx \frac{S_{max} - S_{min}}{S_{max} + S_{min}}. \quad (2.1.26)$$

The sinusoidal approximation makes the analysis of the experimental data easier, as it is a rather simple function to fit.

Quantum vs. classical model Compared to the far-field, where the resulting diffraction pattern at a single grating is a pure wave phenomenon, the periodic fringe pattern from the three-grating-setup can also be mimicked assuming a classical model, at least up to a certain degree. If we think about particles moving on straight rays through the gratings, shadowing effects can lead to high contrast fringe patterns similar to those from the quantum model. To distinguish the KDTLI interference pattern from these Moiré fringes, we have to calculate the expected visibility, in the same manner, using the classical phase-space density function instead of the quantum Wigner function. Since only the second grating is involved in the coherent diffraction, we only have to calculate new coefficients for this one and reuse the coefficients for the first and the third grating as well as for the free time evolution. The semi-classical picture of the interaction between polarizable particles can be described by classical momentum kicks due to the optical dipole potential. The coherent phase effect becomes

$$\beta_{1,cl}(\xi) = \phi_0 \pi \xi, \quad (2.1.27)$$

with the contribution of the photon absorption staying the same as for the quantum model. Substituting the Talbot coefficient with the classical coefficient we

can express the visibility as [14]

$$V_{\text{cl}} = 2 \operatorname{sinc}^2(\pi f) \tilde{B}_{2,\text{cl}} \left(\frac{L}{L_T} \right). \quad (2.1.28)$$

A major difference between the quantum and classical prediction is that the contrast of the Moiré-fringes lack periodicity in the Talbot length. We can also take advantage of the specific optical power dependence to differentiate between the quantum and the classical model.

2.1.2 Matter-wave assisted deflectometry

Matter wave interferometers are not only intriguing tools to probe the foundations of quantum physics [2, 3] but they are also a useful platform for quantum assisted molecular measurements [8, 9, 106–109]. The possibility to write and read interference fringes on the nanoscale allows measurements of tiny shifts caused by forces acting on the delocalized molecules. The ideas of matter-wave assisted deflectometry builds on early beam deflection measurements with clusters [110–113] and molecules [114, 115]. Early metrology experiments with matter waves, measuring the photon recoil on Cs atoms [116], static polarizabilities [117, 118], the gravitational acceleration [119, 120] and the earth's rotation [121, 122] followed by many more interferometric metrology experiments measuring electric [12, 123, 124], magnetic [7, 11], structural [125, 126] and optical [127–129] properties.

Effects of forces on the interference fringes If we follow up on our previous calculations using the phase-space representation, a force that acts laterally on the delocalized particle and causes a shift of Δx has the following effect on the transmission function

$$\tilde{S}(x_S) = S(x_S) \exp \left(i \frac{2\pi}{d} (\Delta x_1 - 2\Delta x_2 + \Delta x_3) \right), \quad (2.1.29)$$

with Δx_i the equivalent shift of the i^{th} grating with respect to the particle beam [130]. While for monochromatic beams this results in a shift of the interference fringes, a finite velocity spread additionally leads to phase averaging and thus to a reduction of the fringe visibility. Since both contributions carry information on the deflection process, both can be used to draw conclusions about the acting forces or the molecular properties if the perturbations are well characterized. In the interferometric setup, the precision does not depend on the shift of a single beam spot but on many interference fringes. A major advantage over classic beam deflection measurement is the gain in accuracy, considering the small period of the fringes (266 nm at LUMI) and the ability to resolve the shifts on a nanometer-scale.

Inertial forces We differentiate between two types of measurements: The measurement of inertial forces and the measurement of internal molecular properties. What inertial forces like gravity or rotations have in common is that they affect the particles throughout the entire length of the interferometer. While significant advantages in inertial sensing have been reported in atom interferometers [131–133], LUMI is less well-suited for this application, especially with large molecules. The high precision of the atom interferometers are mostly due to the good signal-to-noise ratio. At LUMI, difficulties regarding the beam flux and the overall interferometer stability arising with large molecules make it challenging to compete with modern atom interferometers. Nevertheless, due to the long baseline, LUMI is still so sensitive to inertial forces that the effect of Coriolis force on the interference fringes has to be compensated [10].

Molecular properties We can take advantage of the high sensitivity in combination with the universality of LUMI. The ability to operate with complex molecules makes our interferometer well suited to measure molecular properties. Different classes of particles can be compared with each other using the same setup. The internal molecular properties can couple to the transverse motion when the interfering particles are exposed to external fields. By measuring the resulting shifts or the reduction of contrast, the internal properties

can then be evaluated. A magnetic field can enable access to the magnetic susceptibility and similarly an electric field to the static polarizability. The key point to exploit the full potential of the sensitivity is to know the external fields as well as possible. A more detailed discussion of the effects on the interference due to a magnetic and electric deflection in an external field is given in section 3.2.3 of the following chapter.

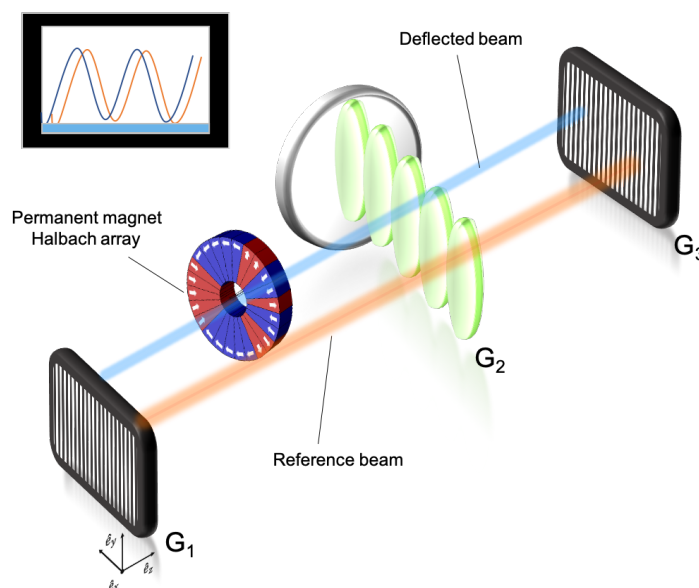


Figure 2.7: Schematic illustration of an interferometric magnetic deflectometry measurement. The deflecting magnet is placed in front of the second gratings to increase the effect of the shift onto the interference fringes. Similarly to a classical beam deflection experiment the whole molecular beam is shifted in the TLI setup with a major difference that we can measure the shift of the interference fringes rather than of the single beam. This leads to an increased accuracy over classical beam deflection experiments.

2.2 Macroscopic quantum effects

How come we can observe quantum behavior for electrons, atoms and even large molecules within the interferometer but the macroscopic objects in our

everyday life do not show any sense of wave properties? Is it just the mass or the size of the object that suppresses the wave behavior or is it something else? Matter-wave interferometry cannot give a final answer to this but so far there has been no experimental evidence a mass or size-related boundary. Even though that we have demonstrated quantum superposition of molecules with masses up to 28.000 amu [15], there is still a lot of work ahead to push the limits of the macroscopic matter-wave experiments towards even more massive and more complex systems.

Superposition in the interferometer Considering that the molecules from our experiments are internally hot, there is an immense amount of different vibrational and rotational states that are occupied within the ensemble. This means that the molecules are quantum-mechanically not in the same state and therefore distinguishable. However, the cat-states that live in our matter-wave interferometer, only affect the center-of-mass motion of the molecules and all that matters for the outcome of the experiment is the *which-path information*. This explains why we can observe quantum superposition of internally hot molecules consisting of up to 2000 atoms [15]. At this point it should also be emphasized that what we observe in our interferometer is the interference between center-of-mass positions of single molecules. Therefore it makes no difference if the particles are in different internal states, as long as the paths are indistinguishable. If the which-path information remains unknown throughout the experiment, the quantum system will keep its coherence and exhibit interference phenomena.

Decoherence Both of the two most relevant mechanisms that can destroy the interference pattern by decoherence involve a measurement of the particles' position, hence the flow of the which-path information into the environment [134]. One is collisional decoherence, describing collisions of the coherent particle with environmental particles and the other one is thermal decoherence, for which the position can be determined by thermal radiation. In order to lose coherence, the measurement of the position must be on the order of the grating period.

This can for example be a single photon with $\lambda_\gamma \leq d$ or many photons with a longer wavelength. Except for experimental conditions with isolated particles, everything in the real world is usually embedded in the environment and all the particles are frequently colliding with each other. Considering this, it becomes obvious, that due to decoherence, no macroscopic quantum effects will be seen in our everyday world [135].

Interpretations of quantum mechanics Decoherence theory explains the appearance of classical behavior by applying the rules of quantum mechanics but it doesn't address all philosophical questions. There have been several different approaches on how to describe and interpret the transition from quantum mechanics to the classical world (fig. 2.8). In the early ages of Niels Bohr and Heisenberg, physicists suggested a strong distinction between quantum-mechanical and the classical world [136]. However, they never referred to any discrete boundary, but ascribed the transition to measurement, which is known as the Copenhagen interpretation [137]. Others believed that it takes a conscious observer measuring for the wave function to collapse [138]. If you want to omit the measuring process as a whole, you can for instance follow Everetts many-world interpretation [139], where he suggests that every possible quantum outcome takes place in a different world. Another group of interpretations are objective collapse models [140]. These models suggest a modification of quantum theory by adding a non-linear term to the Schrödinger equation. This additional term then leads to the collapse of the wave function independently of any observer. Well-known examples are Diosi's [141] and Penrose's [142] idea of the gravitationally induced collapse and the Ghirardi-Weber-Rimini theory [143], with an adapted version known as the continuous spontaneous localization theory (CSL) [144].

2.2.1 Continuous spontaneous localization

While these quantum interpretations are difficult if not impossible to test or distinguish in the laboratory, the CSL theory provides experimentally accessible

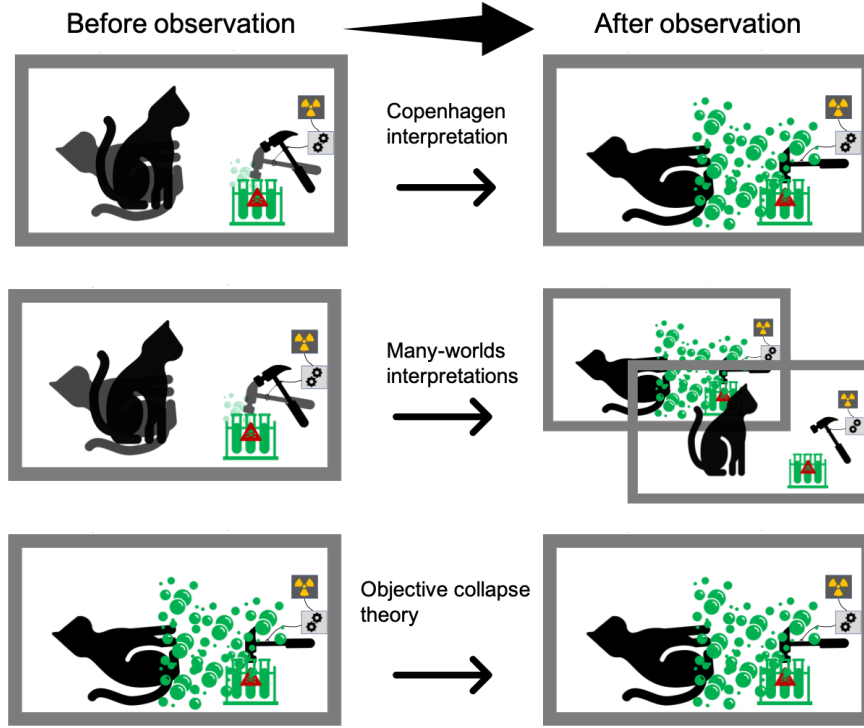


Figure 2.8: A schematic illustration of the different quantum interpretations based on the example of Schrödinger's cat. Starting with the Copenhagen interpretation, the cat is in superposition of dead and alive before opening the box. As soon as the measurement takes place, the wave function collapses into one or the other state. According to the Copenhagen interpretation, reality is only created through the measurement process. For the many-worlds interpretations we also start with the superposed cat before observation. However, if a measurement takes place, the world divides into two, so that both results can coexist in each one of the worlds. A different starting position is depicted for the objective collapse theory. Since the collapse of the wave-function is described by a stochastic differential equation, it can take place any time, independently of any observer. So when the box is finally opened, we may see a dead cat but it could be, that the cat was already dead before we observed it.

parameters. The theory suggests a stochastically driven collapse of the wave function to a localization radius r_c and at a certain collapse rate λ_{CSL} [140]. These two constants depend on a coupling constant γ ,

$$\lambda_{CSL} = \frac{\gamma}{(4\pi r_c^2)^{3/2}}. \quad (2.2.1)$$

and build the key elements of the theory. Especially the quadratic scaling with mass makes matter-wave interference with high masses a viable tool to test the CSL theory. The superposition states can help to set bounds for the collapse rate λ_{CSL} and the localization rate r_c . The two values correspond to the coherence time, i.e. flight time through the interferometer and the length scale, i.e. the spatial extent of the superposition. A closed expression, connecting the fringe visibility of a Talbot-Lau matter-wave interferometer with the localization parameters λ_{CSL} , r_c has been derived as

$$\frac{V_{CSL}}{V} = \exp \left\{ -2\lambda_{CSL}\tau \frac{m^2}{m_0^2} \left[1 - \frac{\sqrt{\pi}r_c}{N_T d} \operatorname{erf} \left(\frac{N_T d}{2r_c} \right) \right] \right\}, \quad (2.2.2)$$

with the reference mass $m_0 = 1$ amu and τ the flight time of the molecules [145]. The superposition observed in our experiments can then exclude pairs of parameters that predict a contrast loss of $V_{CSL}/V = 0.5$ whenever we observe 50 % of the expected visibility.

2.2.1.1 Macroscopicity

A generalized measure of how macroscopic a superposition state is, was introduced by Nimmrichter and Hornberger [146]. The macroscopicity μ quantifies the extent to which the newly gained boundaries exclude modifications of quantum theory and is well suited for comparing different quantum systems [15, 93, 133, 147]. The definition for the macroscopicity is given by

$$\mu = \log_{10} \left(\frac{\tau_e}{1s} \right) \approx \log_{10} \left(\left| \frac{1}{\ln f} \right| \left(\frac{m}{m_e} \right)^2 \frac{\tau}{1s} \right), \quad (2.2.3)$$

with τ_e as the largest excluded time parameter, f the fidelity of the interference pattern and m_e the electron mass. This macroscopicity compares the experiment to the superposition of an electron during a certain time scale τ . A macroscopicity of 14 corresponds to an electron being in a superposition

state over a period of 10^{14} s. The highest interferometric macroscopicity was recently set at LUMI by observing a superposition of molecules with masses up to 28.000 amu [15]. The results leading to this value are discussed in detail in chapter 4. It is worth noting that other macroscopic quantum effects have been observed with matter-wave interferometry [15, 93, 133, 147] but also with SQUIDS [148], BECs [149] and optomechanical systems [150–154], to name a few.

Chapter 3

Long Baseline Universal Matter-wave Interferometer

Molecular matter-wave interferometry is a useful tool to probe the quantum nature of objects. Several experiments have been performed to confirm its validity for even massive and complex molecules [15, 71, 92, 155]. The near-field Kapitza-Dirac Talbot Lau Interferometer (KDTLI) performed particularly well when it comes to high mass matter-wave interference. The former interferometer was able to demonstrate quantum superposition of molecules with masses beyond 10.000 amu [93]. However, the setup of the KDTLI was not built to cope with higher masses. More precisely, the geometrical restriction allows experiments with de Broglie wavelengths ≥ 700 fm, corresponding to a molecule with a mass of 30.000 amu and a center of mass velocity of 19 m/s. In order to allow even heavier molecules to interfere, one could either make them slower or extend the baseline of the interferometer. Suitable sources and gravitational limitations motivate the implementation of the long-baseline universal matter-wave interferometer (LUMI), an upgraded version of the KDTLI, with a 10 times longer baseline compared to the predecessor.

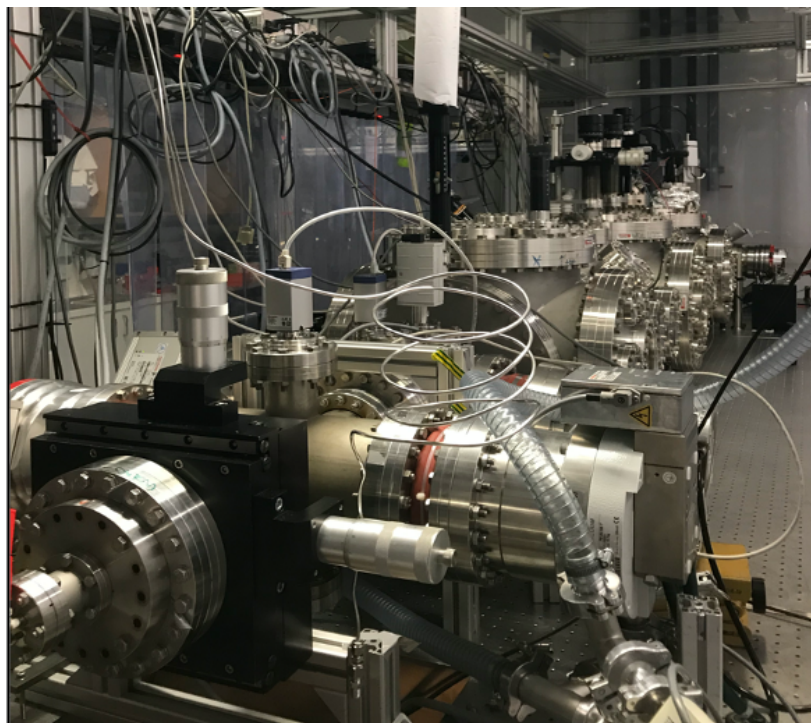


Figure 3.1: A picture of the actual LUMI setup. The whole experiment is built inside connected vacuum chambers which can be seen in this picture. The vacuum chambers take up almost the entire length of the five-meter long optical table. Thinking of the size of the molecules and the interference pattern we experiment with, the vacuum chambers may seem oversized. The large chambers are nonetheless needed if you consider which parts have to be included for the experiment: The molecular source, differential pumping stages, a velocity chopper disc, collimation slits, a two-meter long and 140 kg heavy Invar bar with several vibrational damping systems, the interferometer, two quadrupole mass spectrometers, deflection electrode and magnets and coils, among other components.

Experimental Setup

LUMI, like its predecessor, is a three-grating near-field matter-wave interferometer. The baseline of the interferometer is two meters long, which makes LUMI ten times longer than our former near-field interferometer. The first and the third grating (G_1 & G_3) are nano-fabricated SiN gratings with a period of $d = 266$ nm. The second grating (G_2) can either be another material grating with the same specifications as G_1 and G_3 or realized by a standing

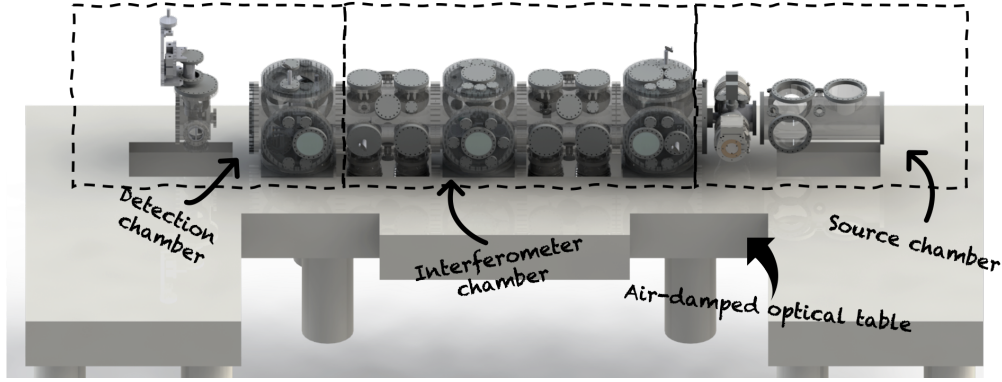


Figure 3.2: A Solidworks drawing of the interferometer which is resting on an air-damped optical table. The setup can be mainly divided into three individual sections: The source chamber, the interferometer chamber and the detection chamber. All chambers are connected via gate valves and can be pumped and vented individually.

lightwave matching the period of the other gratings ($\lambda_{\text{Laser}} = 532 \text{ nm}$). The inter-grating separation is $L = 0.98 \text{ m}$ and the interferometer operates in the symmetric configuration with $L_1 = L_2 = L$ and $d_1 = d_2 = d_3 = d$. The MPM (material, optical phase grating, material) scheme with a period of $d = 266 \text{ nm}$ is suitable for de Broglie wavelengths as low as $\lambda_{dB} = 35 \text{ fm}$ (fig. 5.2) [6]. The whole setup is built inside vacuum chambers that are resting on a 5.5 m long optical table. Low pressures in the range of $\approx 10^{-9} \text{ mbar}$ (UHV) are necessary to avoid collisional decoherence within the interferometer. The wide sidearms of the table allow a lot of space for additional devices such as laser systems or molecular beam sources.

The experimental setup can be coarsely separated into three individual sections: the source chamber, the interferometer chamber and the detection chamber. While housing different parts of the experiment, the chambers are all connected via vacuum gate valves. This allows, each section to be pumped and vented individually, with separate backing pumps responsible for each section. These pumps are located in an adjacent room to vibrationally decouple from the interferometer.

3.1 Source chamber

The source chamber is composed of the molecular beam source and a differential pumping stage. The source is pumped by three turbomolecular pumps (TMP, 700 l/s). Another TMP (700 l/s) ensures the differential pumping stage, resulting in a total of four vacuum pumps for this part of the interferometer. However, due to the gas load of the molecular sources, the pressure inside the chamber is usually in the range of $10^{-8} - 10^{-7}$ mbar, which is two orders of magnitude higher than the UHV inside the interferometer chamber. The differential stage helps to avoid contamination of the entire interferometer with source material and to keep the pressure low in the adjoining chambers. The high number of pumps is not only needed to counteract the high pressures that can be generated by some of the sources but also to get back swiftly to sufficient low pressures after venting the chamber. This is frequently the case whenever the chamber has to be opened to refill the sources or when swapping to a different molecular beam source.

3.1.1 Molecular beam sources

Since the beginning of molecular matter-wave interferometry sources have always been a challenging, if not the most challenging, part of the experiments. The long-baseline interferometer is able to perform interference experiments for a very diverse class of particles, ranging from atoms to biomolecules and soon also large metal clusters. These different types of particles come with a large variety of internal properties, thus making a selection of different molecular beam sources essential. Although the Talbot-Lau scheme comes with relaxed coherence requirements, some important properties for the molecular beam source must be considered to be compatible with our experiments. When the molecules pass the interferometer, they should preferably be intact, slow, sufficiently cold and neutral. The source must furthermore generate a stable beam of sufficient flux into a well-defined direction. Despite these strict requirements several sources are available that work for matter-wave interferometry.

Effusive beam source

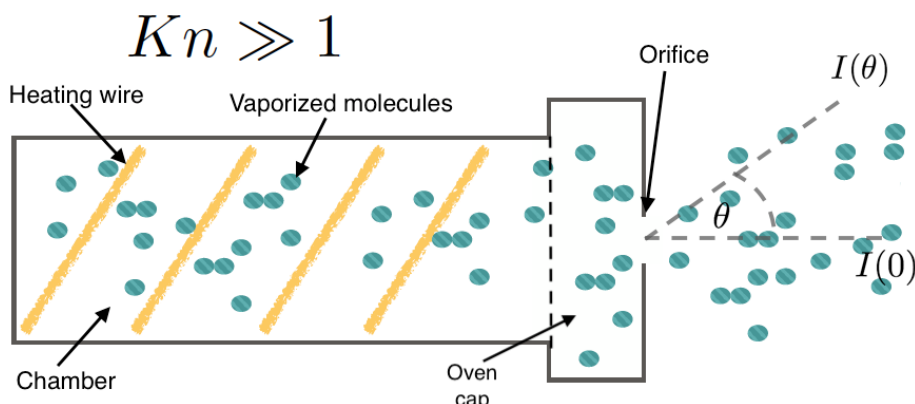


Figure 3.3: Schematic diagram of an effusive cell. The oven cylinder is filled with the molecules which are usually dissolved or in the form of a powder. A high resistance wire is wrapped around the cylinder body which allows to heat the oven electrically. The temperatures can easily reach up to 1000 K when the cylinder is covered with a thermal shielding. We usually monitor the temperature with a thermocouple (type K) placed in the cylinder wall. The diameter of the orifice is in the range of 100 – 500 μm . To obtain an effusive beam with a Maxwell-Boltzmann velocity distribution, the Knudsen number must be $Kn > 1$, which means that the mean free path of the molecules inside the cylinder is larger than the dimensions of the orifice.

The simplest version of a molecular beam source is a closed cylinder wrapped in a heating wire with a small slit in the cap, a so-called Knudsen cell. The molecules inside the cell are heated to sublimation and effused through the orifice in the cap. To obtain an effusive beam, i.e. a molecular flow, the mean free path Λ of the molecules has to be larger than the smallest representative dimension of the orifice d^1 . The ratio Λ/d is also called the Knudsen number Kn [156]. The parameter helps to distinguish between different flow regimes of the fluid dynamics (fig. 3.4). The mean free path is further defined as the distance that the molecule travels between two collisions given by $\Lambda = k_B T / (\sqrt{2} p \sigma^2)$, with k_B as the Boltzmann constant, T the temperature, p the vapor pressure and σ the molecular diameter.

¹This can correspond to various physical traits of a system. In this context it is mostly related to the aperture size or the nozzle length.

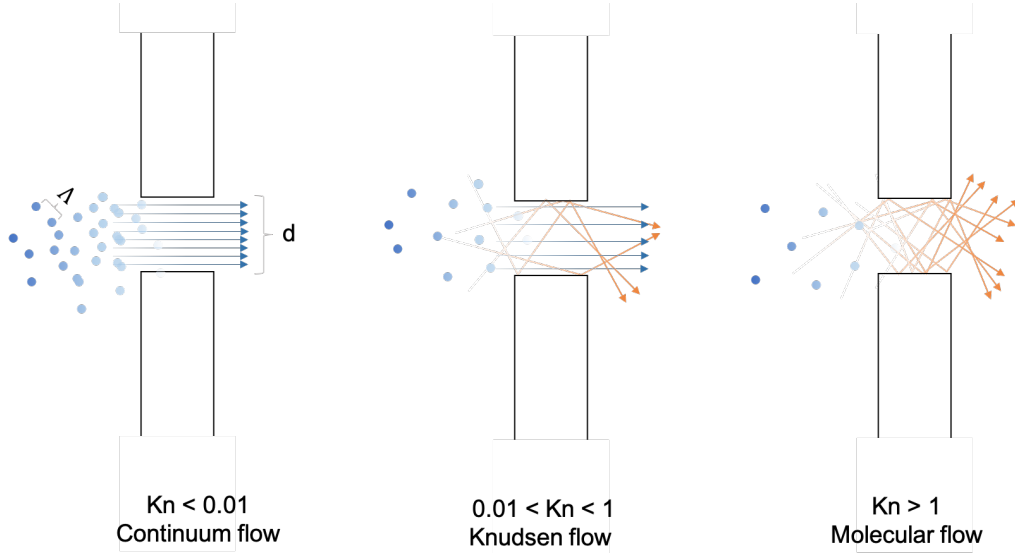


Figure 3.4: A schematic diagram showing the different possible fluid dynamics using the Knudsen number as the parameter. When the mean free path is much smaller than the orifice ($Kn < 0.01$), the particles will collide frequently and form a directed continuous flow. The dynamics are then described by the laws of continuum mechanics. On the opposite, when the mean free path is large compared to the orifice ($Kn \gg 1$), there will be almost no collisions. The dynamics of the molecules can be treated on a molecular level, leading to the scope of statistical mechanics. Everything in between is usually called transitional or Knudsen flow.

The fluid dynamics of a Knudsen cell are covered by the theory of statistical mechanics and the velocity distribution can be described by the Maxwell-Boltzmann law

$$f(v) = \frac{4}{\sqrt{\pi}} \left(\frac{m}{2k_B T} \right)^{3/2} v^2 \exp\left(-\frac{mv^2}{2k_B T}\right), \quad (3.1.1)$$

with m as the molecular mass and v the forward-directed center-of-mass velocity. The mean velocity of the particles is given by $\hat{v} = \sqrt{2k_B T/m}$ and the beam comes with a broad $\cos(\theta)$ -distribution (fig. 3.3). The intensity in number of particles per unit solid angle and unit time is given by $I(\theta) = I(0)\cos(\theta)$ with

$$I(0) = \frac{p(T)\hat{v}A}{k_B T 4\pi} \quad (3.1.2)$$

and θ as the angle between the solid angle and the normal of the orifice plane and A the orifice area through which the particles leave the source [157]. To maintain a molecular flow it is also important to avoid collisions with residual gas particles by keeping the pressure low in front of the nozzle.

The Knudsen cell is simple and provides a stable continuous molecular beam. It has therefore often been the first choice for matter-wave experiments in the past [4]. However, not all types of molecules are compatible with ovens, since they should not fall apart during the heating. This mostly holds for molecules up to 1000 amu but as larger molecules exhibit more complex structures, fragmentation is mostly unavoidable, especially when it comes to biological molecules. Tailoring the molecules with fluoroalkyl-chains can help to increase the volatility by reducing the enthalpy [158]. It is also beneficial regarding high-mass interferometry since the chains increase the mass to polarizability ratio and add to the total mass. Using this technique the former KDTLI experiment was able to perform matter-wave experiments, using a thermal beam of molecules with masses exceeding 10.000 amu [93].

Although the scope of the thermal oven is limited to atoms and thermostable molecules it is a very useful source even for high-mass matter-wave interferometry. At LUMI we have used thermal ovens to align and calibrate the interferometer with C_{60} or Cs atoms throughout several experiments [11, 15, 159].

Supersonic beam source

One can also use the source cell to produce supersonic jets by increasing the pressure inside the crucible. The underlying idea is to release the particles from a high-pressure reservoir into a low-pressure background through a small hole or nozzle [161]. The short mean free path length causes the particles to collide

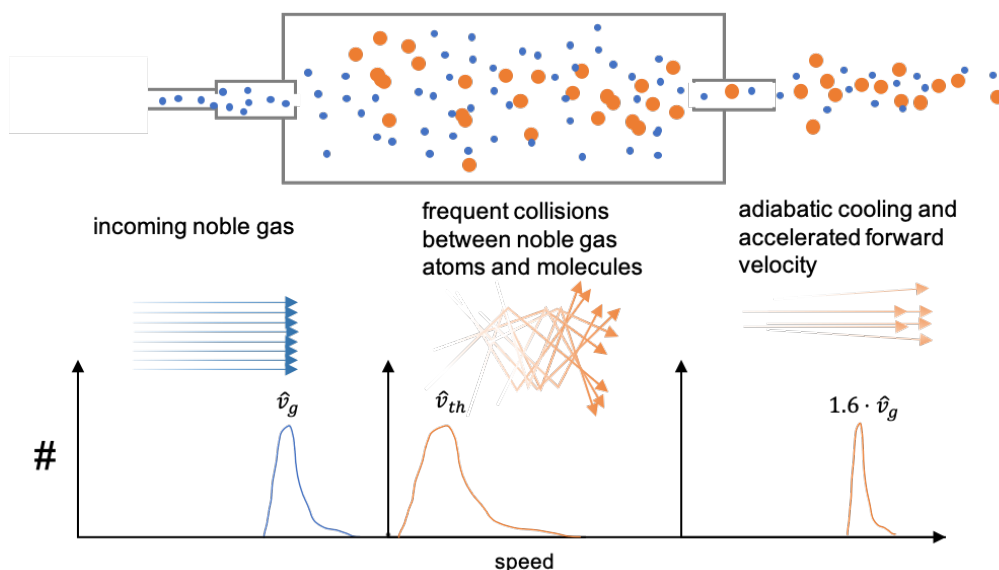


Figure 3.5: Schematic operating principle of a seeded supersonic beam source. The noble gas flows into the oven cell, which is filled with the vapor of the molecules. The gas atoms collide with the molecules and accelerate them approximately to the velocity of the noble gas atoms v_G . During their escape through the nozzle, the molecules are colliding frequently with each other and the nozzle walls. The collisions convert the translational energy into an acceleration of the particles in the forward direction and the adiabatic expansion additionally cools the molecules during their escape. The resulting velocity spectra of the molecules is narrower and shifted to a higher mean velocity of approximately $1.6 \cdot v_G$ for monoatomic noble seed gases, compared to the thermal beam [160].

frequently during their escape, converting most of the energy into the forward-directed and accelerated beam. The fast expansion of the particles also provides adiabatic cooling within the beam. The dynamics of supersonic expansions are described by the laws governing a continuum flow (fig. 3.4).

A common way to generate supersonic beams of molecules is seeding in a monoatomic carrier gas (fig. 3.5) [162]. High pressures are easily obtained by mixing the molecules of interest with a light diluent gas, usually a noble gas which flows into the crucible under high pressure. The molecules thermalize with the noble gas and the mixture is accelerated by the adiabatic expansion to a velocity of about $1.6 \cdot v_g$ in the case monoatomic noble seed gases, with

v_g the most probable thermal velocity of the gas [160]. The velocities are usually in the range of 300 m/s for cryogenic nozzles and can exceed 1000 m/s at room temperature. The fast expansion also provides adiabatic cooling for the translational, rotational and vibrational degrees of freedom, ranking $T_{trans} < T_{rot} < T_{vib}$ [163]. The resulting velocity distribution is narrowed and at the same time the maximum shifts towards higher velocities (fig. 3.5) compared to the thermal beam. The final beam properties strongly depend on the carrier gas type, background pressure, nozzle geometry, the mass and the heat capacity of the seeded molecules [160].

High gas loads of the carrier gas may lead to a significant increase in the pressure inside the source chamber. This can on one hand be a problem for the vacuum pumps and on the other hand cause unwanted collisions and turbulences within the molecular beam. The total gas load can be reduced by pulsing the nozzle, for example using an Even-Lavie valve [164].

However, also the seeding technique requires one to bring the desired molecules into the gas phase to generate a molecular beam. Unfortunately the application of thermal vaporization is limited to thermostable molecules and for large, complex and particularly biological molecules heating usually leads to significant fragmentation. The use of cold noble gases and also the adiabatic cooling can help to distribute the heat energy before the molecules fragment. Nonetheless, the small de Broglie wavelengths that come with the high velocities make the source impractical for high mass matter-wave interferometry. On the other hand it can be helpful in specific metrological measurements, as it can be necessary to reduce the interaction time of the molecules with the external fields or with the gratings.

Laser-induced desorption source

A promising approach to bring fragile and heavy molecules into the gas phase is laser-induced desorption (fig. 3.6). Tightly focused nanosecond laser pulses are used to minimize the thermal fragmentation. This strategy is widely used

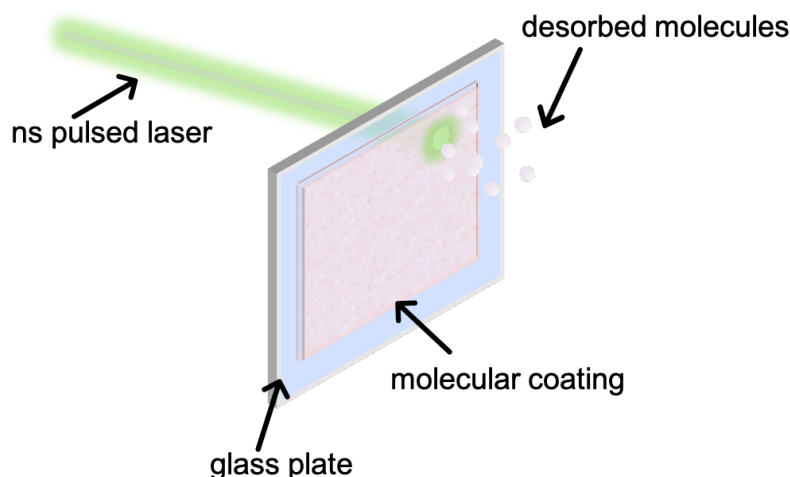


Figure 3.6: Schematic diagram of a laser desorption source. A glass plate is coated with the molecules and a nanosecond-pulsed laser beam is focused onto the backplane of the plate. A possible explanation for the desorption is that the nanosecond pulses are absorbed by the molecules, while heating the sample locally with a heating rate of up to 10^{11} K/s. This is enough to overcome the binding energies and bring the molecules into the gas phase. Due to the low heat load of the short pulses, the molecules tend to remain intact during their takeoff.

in the field of mass-spectrometry known as matrix-assisted laser desorption and ionization (MALDI). In MALDI the objective sample is mixed with a suitable matrix, which is responsible in absorbing the energy from the laser pulse. The absorption further triggers the desorption of the matrix-sample-mixture. This soft-desorption/ionization technique works for complex biomolecules and massive proteins up to 10^6 amu without significant fragmentation [165]. MALDI is intended to produce beams of ions rather than of neutral molecules, which is indeed preferable for mass analysis but impractical for matter-wave interferometry. In order to get around a post neutralization scheme one can use matrix-free laser desorption (LD), which has shown to produce neutrals to a large extent [166]. Since the sample itself has to absorb the light is not as soft as MALDI. The damage can be reduced to a minimum, by using very short intense pulses of light. While it is not fully clear what mechanisms are involved in the desorption process, the molecular beam properties depend strongly on

the laser parameters, the molecular mass of the sample and the quality of the coating.

3.2 Interferometer chamber

The interferometer chamber is the heart of LUMI. It consists of three connected vacuum chambers (CF 350) which house the vibrationally isolated interferometer with the three gratings as well as various deflection systems. The chambers are pumped by two ion getter titanium sublimation pumps and two TMPs (700 l/s), allowing a base pressure of about 2×10^{-9} mbar.

3.2.1 Gratings

The gratings are supported by six-axis motorized stages (Smaract) which are mounted on a 140-kg heavy Invar bar, forming the 2 m baseline of the interferometer. The slip-stick motors allow translational and rotational displacements over a large travel range with a nanometer accuracy in translation and an increment of 15 microdegrees for the rotational axes. All three gratings can be moved out of the beam completely. This is necessary to increase the particle flux for beam alignment procedures and to prevent the gratings from clogging by limiting the exposure time to the interference scans. The grating-free beamline also allows us to perform source and detector tests directly in the setup of LUMI.

While the first and the third grating are always nano-fabricated material gratings ($d = 266$ nm), the second grating can additionally be changed in situ to an optical phase grating. For this, the material grating is withdrawn and a 532-nm laser beam (Verdi V20) is focused with a cylindrical lens ($f = 100$ mm) onto a high-reflective 0° mirror. A standing light wave with a period of 266 nm is then formed by the retro-reflected laser beam. The assembly of the second grating has an additional translational stage on which the material grating, the mirror and the lens are mounted allowing us to adjust the standing light wave in situ.

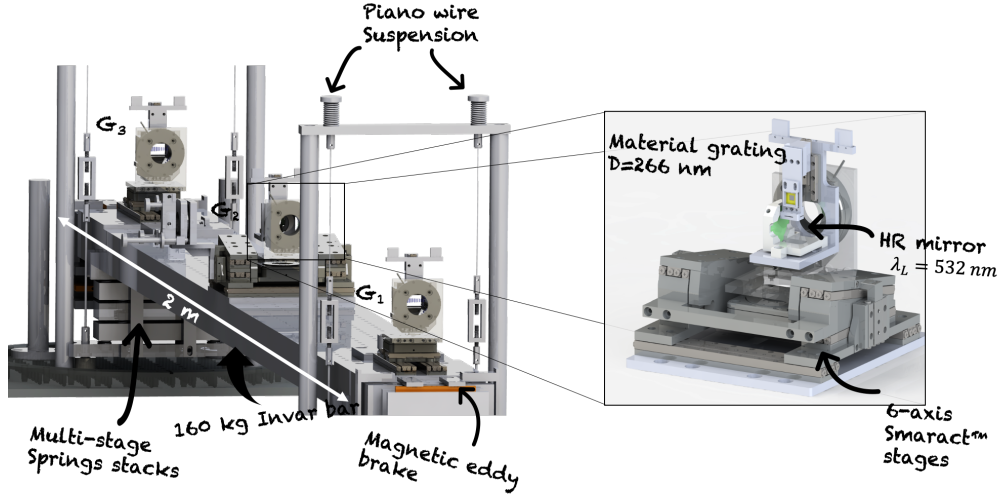


Figure 3.7: **Left:** The 2 m baseline of LUMI and the vibrational damping systems. The 140 kg heavy baseline is made out of Invar to ensure thermal and mechanical stability. The Invar bar is resting on multi-stage spring stacks which are suspended from piano wires. Furthermore, magnetic eddy brakes are mounted at the ends of the bar to damp relative motions w.r.t the vacuum chamber. The only contact between the baseline and the vacuum chamber is via the piano wires. **Right:** The baseline is holding all three gratings which are mounted on six-axis motorized stages. All three gratings can be withdrawn from the beamline. The second grating can be interchanged by an optical grating by moving the material grating out of the beam and focusing a laser beam (532 nm) with a cylindrical lens onto a HR 0° mirror. The assembly of the second grating has an additional large translational stage which is holding an HR 0° mirror and a cylindrical lens ($f = 100 \text{ mm}$), allowing in situ adjustment of the standing light wave.

3.2.2 Vibrational isolation

A major source of contrast loss can be attributed to vibrational dephasing due to environmental influences. That's why vibrations must be suppressed as well as possible. A vertical grating motion has no significant effect on the interference fringes when the gratings are aligned to gravity but we distinguish three types of horizontal grating motion: independent, common and torsional. LUMI has several damping systems to ensure the required vibrational isolation. The three gratings are mounted on an a 140 kg heavy Invar bar, which is suspended from piano wires ($d = 1.6 \text{ mm}$) and supported by a stack of steel

plates, springs and Teflon balls. Additional eddy current brakes at the ends of the bar ensure sufficient damping of relative motions with respect to the vacuum chamber. To limit the maximum loss of contrast to about 5%, the amplitudes of the vibrations shall not exceed 10 nm [104]. A detailed discussion about the vibrational requirements can be found in reference [130] and the characterization of the vibrational damping systems at LUMI in reference [5].

3.2.3 Deflection systems

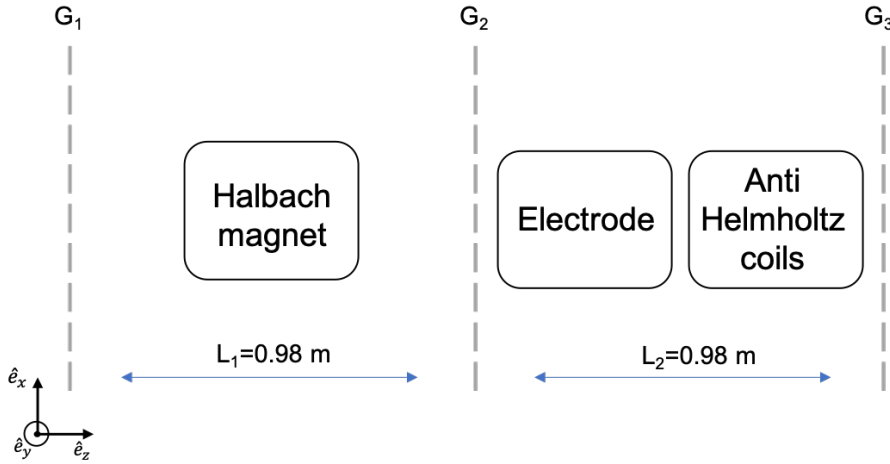


Figure 3.8: Arrangement of the deflection systems installed in LUMI. The permanent Halbach magnet is mounted between G_1 and G_2 , a pair of anti-Helmholtz coils and an electrode between G_2 and G_3 .

The high sensitivity of the interference fringes turns the interferometer into a nano-ruler which can be used for sensitive molecular metrology measurements. Especially the long baseline allows us to sense forces as small as 10^{-26} N. For this purpose the setup of LUMI includes one electric and two magnetic deflection systems (fig. 3.8). The deflection systems can be used to measure static polarizabilities [12], magnetic dipole moments [11] and magnetic susceptibility of the interfering particles.

3.2.3.1 Electric deflection

A specially designed electrode is installed between the second and the third grating to exploit electronic properties of diffracted neutral particles. The geometry of the electrode is designed to have a constant $(\vec{E} \cdot \nabla)E_x$ field and the field strength can be tuned by varying the voltage. The constant field is beneficial to extract the static polarizability from the interference pattern and the fringe shift can be calculated with

$$\Delta\phi = \alpha_{stat} \frac{KV^2}{mv^2}, \quad (3.2.1)$$

where α_{stat} is the static polarizability, V the electrode voltage, m the particle mass, v the center of mass velocity and $K = \frac{2\pi}{d}(-L_1^2/2 - L_1L_2 + L_1L)K_e$ a geometrical factor, with L_1 the electrode length, L_2 the distance between the closest edge of the second grating and the entrance of the electrode, L is the inter-grating separation and K_e the electrode geometry factor. While this factor already includes the spatial deviations from the electric fields within the area of the particle beam, phase averaging due to the velocity spread has to be considered separately. The finite velocity distribution of the particle beam leads to different interaction times for different velocities. The resulting shift distribution washes out the interference pattern. The averaged pattern with amplitude $\bar{A}$ can be calculated by integrating over the velocity distribution $\rho(v)$

$$\bar{A} \cos(kx + \bar{\Delta}\phi) = \int_0^\infty dv \rho(v) A \cos(kx + \Delta\phi) \quad (3.2.2)$$

with the corresponding averaged measured shift being

$$\bar{\Delta}\phi = \int_0^\infty dv \rho(v) A \exp(i\Delta\phi) \quad (3.2.3)$$

with $k = 2\pi/d$ and d the grating period. As long as the individual fringes remain resolvable, we can extract the static polarizability from the corresponding phase

shift of the interference pattern.

3.2.3.2 Magnetic deflection

Analogous to the electric deflection we can apply a magnetic field to measure internal magnetic properties of the particles. LUMI includes two magnetic deflection systems. The first one, a modified Halbach cylinder, is designed to have a constant $(\vec{B} \cdot \nabla)B_x$ field and is located between the first and the second grating. The magnetic field allows us to measure induced magnetic dipole moments, in a manner similar to the induced electric dipole moment. The observable shifts of the interference fringes allow us to directly measure the magnetic susceptibility χ_{mag} and induced dipole moments $\mu_{ind} \propto \chi_{mag}B$ in the absence of a permanent magnetic moment. Due to the $(\vec{B} \cdot \nabla)B_x$ field, a nuclear magnetic moment is already sufficient to reduce the visibility [11]. We can again see the benefits of the constant $(\vec{B} \cdot \nabla)B_x$ field when we write down the force acting transversely on the particles

$$F_x = \chi_{mag} \frac{m(\vec{B} \cdot \nabla)B_x}{\mu_0}. \quad (3.2.4)$$

In the case of a constant force along the flight direction the shift of the interference fringes can be simplified to

$$\Delta\phi = \chi_{mag} \frac{-K(\vec{B} \cdot \nabla)B_x}{v^2\mu_0} \quad (3.2.5)$$

with K again being a geometric factor. In contrast to the electrode K -factor, here the spatial deviations of the fields are not included, which is, together with the lack of oriented magnetic moments with respect to the fields, the reason why the non-uniform position-dependent shifts of permanent magnetic moments will result in a loss of contrast.

Anti-Helmholtz coils

The second magnetic deflection system installed at LUMI is a pair of anti-Helmholtz coils. These coils have been designed and characterized in the interferometer as part of this thesis. The idea behind adding anti-Helmholtz coils was to extend magnetic deflectometry measurements to atoms or molecules with a permanent magnetic moment. While the constant $(\vec{B} \cdot \nabla)B$ field of the Halbach magnet can be used to measure induced magnetic moments, the spatial variation of the field applies inhomogeneous forces to permanent magnetic moments. The tunability and the uniform gradient field of the coils allow us improved control over permanent magnets.

Technical assembly Each coil is composed of two coil frames ($B = 80$ mm, $H = 10$ mm) and a coil body ($A = 65$ mm) which are screwed together (fig. 3.9). A holding block connects the coils to the baseplate ($E = 80$ mm, $F = 160$ mm). Two slit plates, made of a thin steel sheet, are mounted on the entrance and exit. The coil assembly is located between the electrode and the third grating to a 4-axis vacuum manipulator (x, y, z, ϕ) . The coils were wound by hand using a Kapton isolated wire (OD=2.08 mm). Each coil contains a total of 52 turns. The dimension of the coils were designed such that the magnetic field gradient was strong enough to deflect particles with a mass of a few 100 amu and a magnetic moment in the order of a Bohr magneton μ_B , while being able to observe revivals of the contrast. The heat load produced by the currents must remain small enough to operate the system in vacuum without additional cooling systems. The baseplate and the connecting blocks are made from aluminum instead of copper as a compromise between weight and heat conductivity. We made some temperature tests with currents up to 20 A to ensure thermal stability of the assembly. Nevertheless, the coils heat up during operation and the amount of outgassing can get significant for currents above 16 A. This can lead to a reduction of the interference contrast due to collisional decoherence which has been the case during the deflection of cesium (see section 4.2.2).

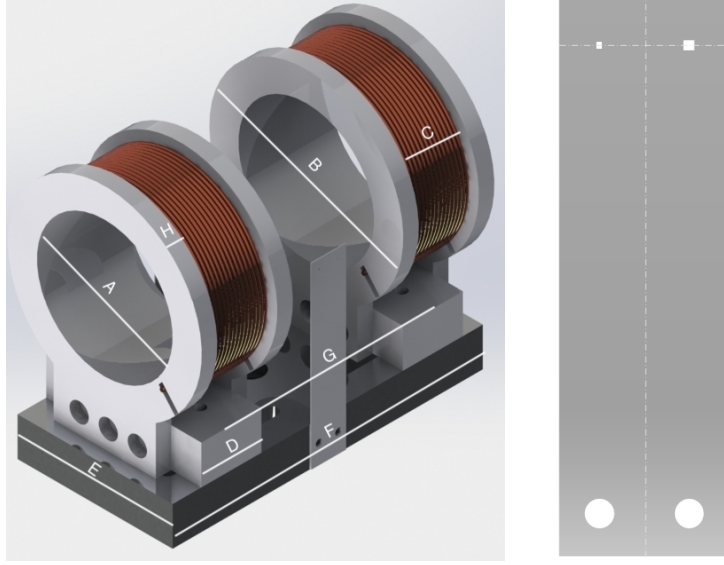


Figure 3.9: Left: Photorealistic view of the anti-Helmholtz coil assembly. All parts except the Kapton isolated wire and steel slit-plates are made of aluminium. The wire was coiled in 4 layers, with 15 turns in the bottom layer, 13 turns in the second and 12 turns for the two uppermost layers. The total resistance of the coils is $R_{T_{20}} = 0.28 \, \Omega$. The distance between the center of the coils (G) is 70 mm ($d \approx \sqrt{3}R$). **Right:** The slit plate has two different sized rectangular slits. The smaller one on the left side has dimensions of $0.5 \times 0.5 \, \text{mm}^2$ and the larger one $1 \times 1 \, \text{mm}^2$. They are both located at the same height of 80 mm and 5 mm left and right off the center.

Magnetic field The anti-Helmholtz coils generate a magnetic field with constant field gradient in the center. Given the force acting on a permanent magnetic moment μ

$$F = -\nabla(\vec{\mu} \cdot \vec{B}) \quad (3.2.6)$$

we can calculate the fringe shift for particles with mass m and velocity v by

$$\Delta\phi(v) = \frac{2\pi}{d} \frac{1}{mv^2} \left[\int_0^{L_1} \int_0^z dz dz' F(z') + L_2 \int_0^{L_1} dz F(z) \right], \quad (3.2.7)$$

with L_1 the travel distance inside the coils and L_2 the distance from the coil

exit to the edge of the third grating. According to Biot-Savart, the field along the coil axis in the center of coils with equal turns N and opposite directed current I can be calculated by

$$B(x) = \frac{\mu_0 NI}{2} \left(\frac{R^2}{(R^2 + (x - d/2)^2)^{\frac{3}{2}}} - \frac{R^2}{(R^2 + (x + d/2)^2)^{\frac{3}{2}}} \right), \quad (3.2.8)$$

with μ_0 as the vacuum permeability, R the radius of the coils, d the distance between the coils and x the position along the coil axis. The most homogeneous gradient field, where the second and the third derivative vanish, can be achieved by choosing $d = \sqrt{3}R$. In this case the gradient in the center is $dB/dx \approx 0,64\mu_0 NI/R^2$. The maximum gradient with $dB/dx \approx 0,85\mu_0 NI/R^2$ can be reached by choosing $d = R$. In both cases the magnetic field centered along the molecular axis ($x=0, y=0, z$) is zero. Since the particles are not pre-selected regarding the direction of the spin, we decided to guide the beam through an area slightly off-center. This way the particles always see a non-zero magnetic field with no abrupt changes of direction. This allows the magnetic moments to align adiabatically with the field and prevent random spin flips.

To obtain the complete 3-dimensional off-axis magnetic fields we solved the elliptical equations derived from Biot-Savart's law numerically (see chapter 4). We used these fields to calculate the interference fringe shifts in dependence of the path the molecules take through the coils. To ensure that the particles pass the coils at the right position we included two slit-plates to the assembly (fig. 3.9).

Effect on permanent magnetic moments The coils allow us to tune the magnetic field gradient and therefore the deflection of the particles by tuning the current. Depending on the direction of the magnetic moment, the particles are shifted in different directions. Together with the finite velocity spread, this leads to different shifts of the interference pattern and therefore to a reduction in visibility. Due to the discrete number of the magnetic moments, the contrast

should recur whenever the shifted interference fringes re-phase (fig. 3.10). The coils were designed such that for small molecules and atoms with a magnetic moment on the order of a Bohr magneton μ_B and a thermal velocity distribution we should be able to observe revivals of the contrast.

We have modeled the expected interference contrast of a deflected particle ($m = 720$ amu, $v = 600$ m/s, $\Delta v = 90$ m/s) with only two magnetic projections, either $+\mu_B$ or $-\mu_B$, as a function of the current to demonstrate the contrast loss and the appearance of the revivals. Averaging over the velocity distribution (blue envelope) leads to a reduction of visibility and suppresses the revivals at higher currents. In principle we should be able to resolve at least the first three revivals in this example. A proper calculation of the shifted fringes includes the hyperfine states and the energy splitting of the magnetic levels due to the external magnetic field of the coils. A precise example of deflected cesium atoms, including all 16 magnetic states is presented in the experimental results in chapter 4.

3.3 Detection chamber

The detection chamber consists of a large vacuum chamber (CF 350) and two separate quadrupole mass selection (QMS) stages. While the common chamber is pumped by a TMP (300 l/s) which is connected to the interferometer chamber via a gate valve, the vacuum chamber housing the high-mass QMS is pumped by an additional TMP (700 l/s).

We usually record our interference scans by measuring the particle flux $S(x_s)$ at the position of the third grating, which acts as a transmission mask. The grating is furthermore scanned transversally (x_s) to reveal the periodic pattern. Since the spatial resolution is limited by the slits of the gratings, it is sufficient to integrate the particle counts as a function of the transversal grating position. In order to make sure that the molecules are intact and also to increase the contrast, the molecules are mass-selected before we detect them. The uni-

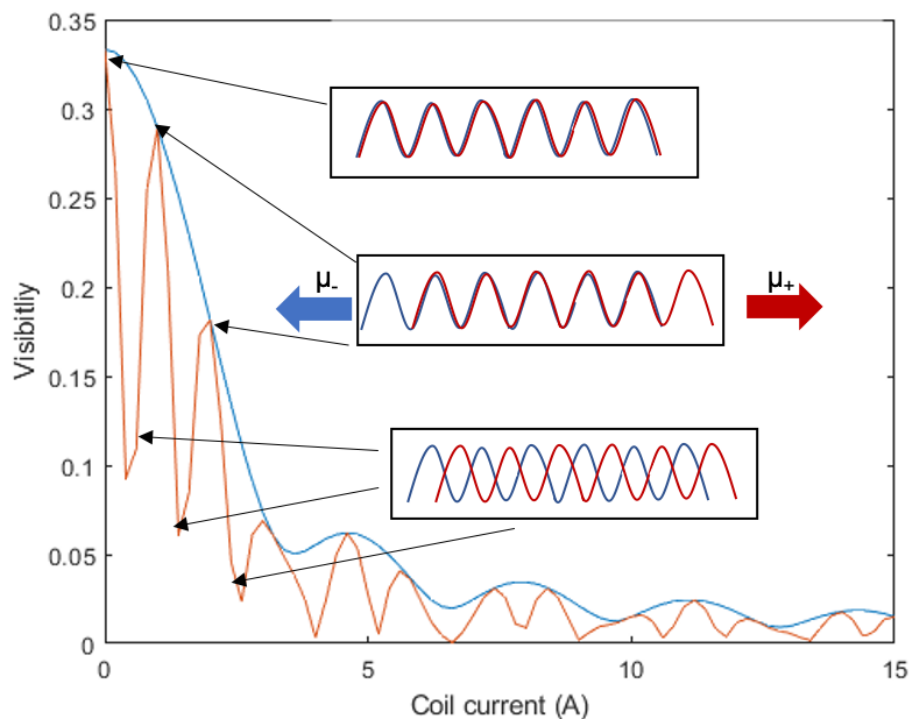


Figure 3.10: Simulated visibility reduction as a function of the coil current, assuming a particle with a molecular mass of 720 amu, a mean velocity of 600 m/s with a 15 % spread and two possible magnetic projections $\pm\mu_B$. The enveloping blue line indicates the loss of contrast due to the velocity spread for a preselected spin. The orange line corresponds to the reduction and revivals of the interference contrast for the shifted magnetic moments, while higher orders are suppressed by the velocity spread. Actually we also have to take the hyperfine structure into consideration, as shown fig. 4.10. However, assuming $s=1/2$ does well in demonstrating how the visibility develops as a function of the coil current.

versality of LUMI is also reflected in the possibility of using different detection schemes. They all have in common that the molecules are ionized, mass-filtered and accelerated onto a dynode. The secondary electrons of the dynode are then accelerated to an electron multiplier where they finally create signal pulses that can be measured electronically.

Mass selection Two mass selection systems are installed in LUMI. One of them is an Extrel MAX-4000HT QMS, which comes with built-in electron bombard-

ment filaments, a conversion dynode and a channel electron multiplier (channeltron). The dynode is connected to a high voltage power supply (10 kV) to improve the conversion efficiency for higher masses. A setup with poles of $d = 19$ mm is compatible with masses up to 4000 amu. It reaches up to 16.000 amu after swapping the poles with smaller rods ($d = 9.5$ mm). For even higher masses we can use a custom-built QMS, consisting of a quadrupole mass filter (QMF) from Oxford Applied Research with $d = 25$ mm rods, a hot filament for electron ionization, an ion guiding assembly from an old Extrel QMS, a dynode and a channeltron. This setup allows us to tune the rf from 3 to 100 kHz while being compatible with masses beyond 10^6 amu.

Ionization technique While in the majority of our experiments we ionize the molecules by electron bombardment, hot wire surface ionization is especially useful for alkali and alkaline-earth atoms with their low ionization energies. The Langmuir-Taylor detector consists of a heated filament of a metal with high work function and works via surface ionization: When neutral atoms land on the wire, there is a certain probability that a valence electron will tunnel from the atom into the material. Due to the high temperature of the wire, the adsorbed atom is released as a positive ion after a residence time, depending on the atomic species, wire properties and temperature, which can range from microseconds to seconds [5]. The efficiency of hot wire ionization depends on the work function Φ_W of the metal and the ionization energy I_e of the particles. The efficiency at a temperature T is given by

$$\frac{s^+}{s^0} \propto \exp\left(\frac{\Phi_W - I_e}{k_b T}\right), \quad (3.3.1)$$

with s^+/s^0 the ratio of ionized to incoming atoms [167]. Langmuir-Taylor detection is more efficient than electron impact, especially for atoms with low ionization energies, such as alkali or alkaline-earths. In LUMI we made use of a hot rhenium wire to ionize cesium, barium and strontium [5].

Chapter 4

Experiments at LUMI

In this chapter, I present matter-wave interference of macroscopic molecules and the interferometric deflection of cesium atoms using the anti-Helmholtz coils. Several more experiments have been performed at LUMI overlapping with the period of this thesis which have already been well documented and published [5, 10–12, 15, 159].

4.1 Macroscopic matter-wave interference

At LUMI we managed to set a new mass record for matter wave interferometry by showing quantum superposition of molecules consisting of up to 2000 atoms [15]. The molecular library includes molecules with masses up to 28.000 amu. This is an increase by a factor of 2.5 and a factor of 10 in macroscopicity compared to the previous interferometric mass record, set by using a magnitude of order shorter near-field matter-wave interferometer [93]. The modified oligoporphyrins (fig. 4.1) were synthesized specifically for our experiment by Patrick Zwick in the group of Marcel Mayor at the University of Basel.

The molecules consist of four zinc-coordinated porphyrins that are arranged around a tetraphenylmethane core. Each one of these branches can hold up to 15 chains perfluoroalkyl chains as denoted by R in fig. 4.1. The functionalization of the porphyrins by adding fluoroalkyl chains increases the volatility of

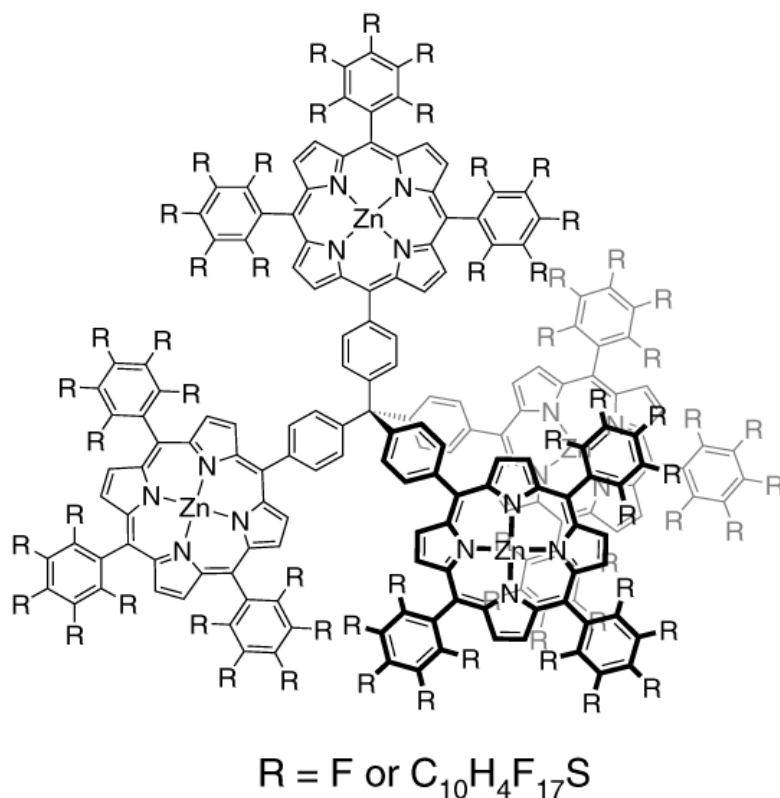


Figure 4.1: Chemical structure of the molecular library that has been used for the high-mass matter-wave interference. The oligoporphyrins were synthesized specifically for this purpose at the University of Basel. The four zinc-coordinated porphyrins are connected via a tetraphenylmethane core. Each porphyrin branch can hold up to 15 fluoralkyl chains, denoted as R . The purpose of the chains is at one hand to increase the volatility but also to increase the total mass of the molecules. Image taken from reference [15].

the molecules, making it easier to form stable particle beams [158]. Another advantage is that the mass of the molecules increases with the number of chains attached to the porphyrins, while exhibiting a higher mass to polarizability ratio. We have used the whole molecular library (fig. 4.2) for interference, with the most abundant mass located at around 26.000 amu. The most massive molecules that we observed throughout the experiment exhibited a mass of 28.000 amu.

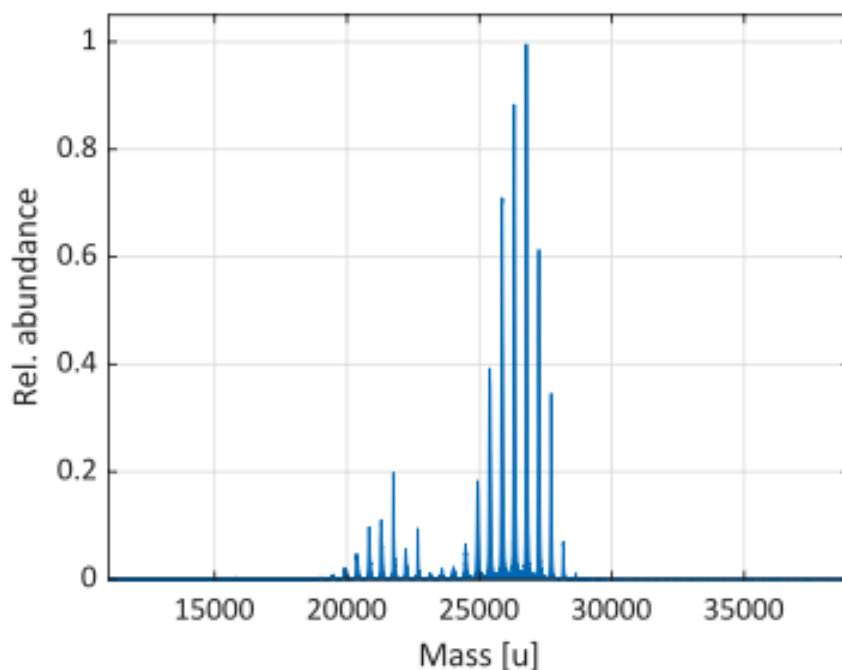


Figure 4.2: Mass spectrum of the oligoporphyrin library that was used throughout the high-mass interference experiment, recorded by Patrick Zwick at the University of Basel. The most abundant mass is located around 26000 amu, corresponding to 50 fluoralkyl chains being substituted. The highest observable mass can be observed at 28000 amu, while containing up to 55 chains. During the interference experiment we have set the QMS to low resolution ($\sim 30\%$) such that the whole library contributed to the interference pattern. Image taken from reference [15].

Experimental setup

Despite the increased volatility of the functionalized molecules, we were unable to create a stable molecular beam using a Knudsen cell. The thermal beam suffered from significant fragmentation and intact molecules were completely absent. Therefore we have opted for a laser desorption source during the further course of the experiment. We dissolved the molecules in *Fluorinert FC-72* and coated a $50 \times 50 \text{ mm}^2$ glass plate (fig. 4.3) with it. The glass plate was screwed onto a frame and the whole assembly was mounted magnetically to a

set of two stepper motors, inside the source chamber. A window at the end of the vacuum chamber gave us optical access for the laser beam. We guided the beam ($\lambda_L = 532$ nm) from a pulsed nanosecond laser (*Edgewave IS120-4L*) through the vacuum laser window and focused it to an area of 0.15 mm^2 onto the backplane of the glass plate. The repetition rate of the laser was set to $f_{rep} = 1$ kHz with an average pulse energy of $PE = 1.8$ mJ during the experiment. This was enough to desorb the molecules directly in the focus spot and its close vicinity. The two stepper motors allowed us to scan the plate in two dimensions, keeping the position of the incident laser, hence the effective position of the source fixed, while constantly resupplying fresh material. The glass plate is shown in fig. 4.3, once after the coating process and once after the molecules were desorbed by the laser.

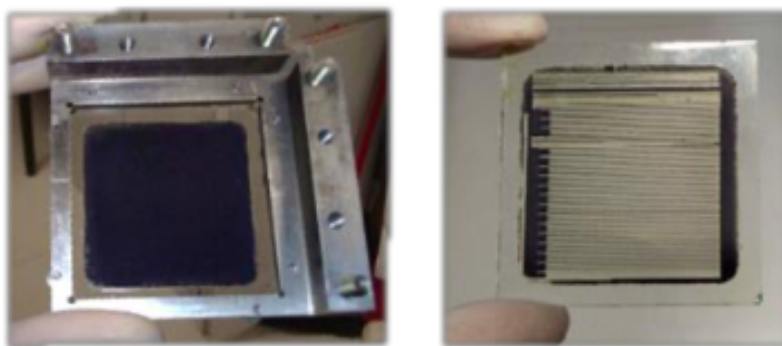


Figure 4.3: **Left:** The glass plate, coated with the oligoporphyrins dissolved in Fluorinert FC-72, mounted onto a metal frame. The picture was taken after the solution has evaporated and right before being mounted onto the magnetic stage. **Right:** The same glass plate after we have used it for our interference experiments. The thin lines that remained in between two desorbed lines were the result of the line separation that we choose to ensure a fresh layer of molecule for every round trip. Photographs taken by Yaakov Fein [5].

Although care was taken to coat the plate as homogeneously as possible, the fluctuation of the molecular flux due to remaining inhomogeneities remained a challenge for the experiment. The count rate typically dropped towards the edge of the glass plate, which is why we avoided this area. This was also the case when the laser shined on the same spot for a longer time, which happened

whenever the stepper motor changed the direction or made a vertical line jump. This is why we only desorbed the molecules in one direction and stopped the interference scans when the motors returned to their starting position. Since the laser did not only desorb molecules in the focal point but also in its close vicinity, we had to set the line separation large enough to supply new material. The fluctuations of the particle counts due to uneven density distribution of the coating made it initially difficult to distinguish the interference fringes from flux fluctuations. In order to solve that, we took reference measurements by mechanically chopping the laser of the optical grating G_2 at a frequency of $f = 2$ Hz. For each point of the interference scans we recorded the particle counts when the laser was blocked and unblocked. By post-analyzing these two datasets, we were then able to filter out the actual interference pattern [5].

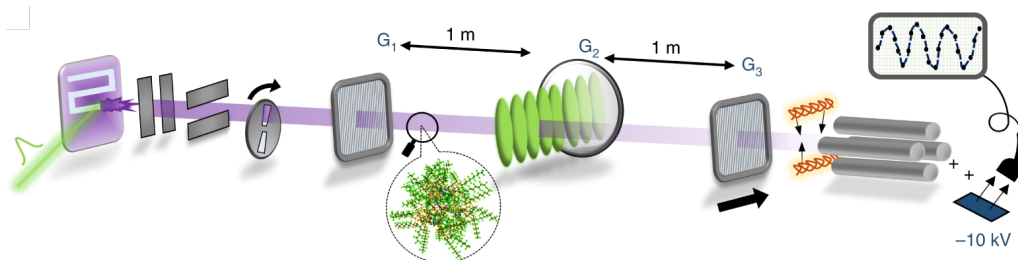


Figure 4.4: Schematic diagram of the setup. The molecular beam was generated by nanosecond pulsed laser desorption ($\lambda_{Laser} = 532$ nm, $f_{rep} = 1$ kHz, $PE = 1.8$ mJ) from a coated glass plate. The beam was then collimated by horizontal and vertical slits (h:500 μ m, v:400 μ m) and a pseudo-random chopper, which allowed us to perform time-of-flight analysis of the velocity spectrum, before entering the interferometer. The interferometer was operated in the KDTLI scheme consisting of two SiN material gratings (G_1, G_3) with a period of $d = 266$ nm and an optical phase grating (G_2) realized by a retro-reflected laser beam (*Coherent Verdi V20*, $\lambda_{Laser} = 532$ nm). The gratings were separated by a distance of $L = 0.98$ m. After passing the third grating, which was scanned transversely across the molecular beam, the molecules were ionized by electron impact, mass-selected with a QMS and detected. Image taken from reference [15].

After the molecules were laser desorbed from the plate, the beam was collimated with horizontal and vertical slits. A pseudo random chopper disc mounted in the source chamber, allowed us to record the velocity distribution of the molecular beam using a time-of-flight analysis [168].

The interferometer was operated in the KDTLI scheme with $G1$ and $G3$ as nano-fabricated gratings with a period of $d = 266$ nm and $G2$ as an optical phase grating, formed by a standing light wave using a 532 nm laser beam (*Coherent Verdi V20*).

We further ionized the molecules by electron impact and selected the masses with a customized QMS (see section 3.3) before the ions were accelerated onto a dynode and detected electronically. Even though this QMS is compatible masses beyond 10^6 amu with a resolution of 2% the low count rates made it necessary to decrease the resolution to about 30 %, detecting the whole mass library (fig. 4.2). While a broadened mass distributions slightly decreases the maximally achievable contrast, the increased count rate and improved signal to noise ratio more than overcompensated this effect. Considering the mass distribution in our calculations (see section 2.1.1), we were able to achieve a high fidelity of 0.93 (fig. 4.5).

Results

To distinguish the interference pattern from classically explainable Moiré fringes, we have recorded the interference scans (see appendix fig. .13) for different optical grating intensities. According to the calculations (see section 2.1.1), the expected visibility as a function of the laser power shows a different behavior for the classical and the quantum model (fig. 4.5), and we can use this power curve to prove the quantumness of our results.

Both simulated power curves are scaled by a factor 0.93 to match our measurement data. The required scaling can be attributed to uncertainties regarding the grating alignment, the opening fractions of the gratings and to a visibility loss due to the Coriolis force. The results are in a good agreement with our expectations and can only be explained by the quantum model. It is also noteworthy that the molecules in our experiment had a mean de Broglie wavelength of about 50 fm. This is five orders of magnitude smaller than the actual diameter of the molecules and seven orders of magnitude smaller than

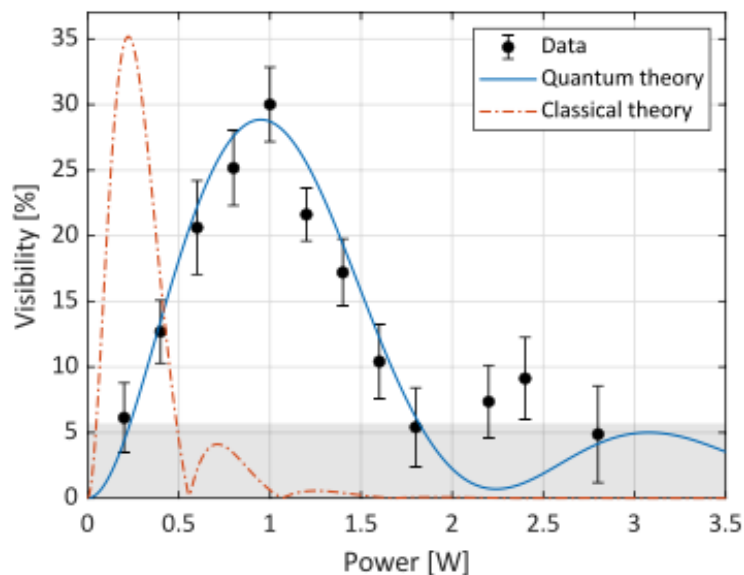


Figure 4.5: Recorded Interference fringe visibility in dependence of the laser power of the optical grating G_2 compared to our calculations. The dashed red line indicates the classical description and the solid blue line the quantum description. Both curves are multiplied with a scale factor 0.93. The measurements results are in a good agreement with the quantum model and cannot be explained by the classical model. Image taken from reference [15].

the minimum delocalization of one grating period. The molecules are in mass comparable to proteins and in number of atoms to small Bose-Einstein condensates (BEC). Yet the molecules are internally hot compared to the atoms of a BEC. Despite the huge number of internally excited states we were still able to observe quantum superposition. The reason is the same as mentioned in section 2.2: Our matter-wave interference relies on the spatial superposition of the centre-of-mass motion of a single molecule and everything that matters is the which-path information.

Macroscopicity and CSL The functionalized oligoporphyrins are the most massive molecules to date for which matter-wave interference could be observed. We can also put this into a more general perspective by calculating the macroscopicity of the superposition using eq.2.2.3. Taking the most abundant mass

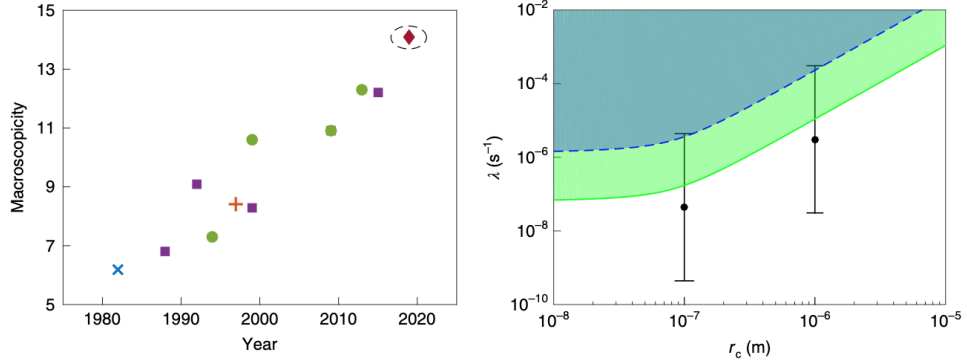


Figure 4.6: **Left:** Macroscopicity values achieved by selected matter-wave experiments over the past years. The circle red diamond corresponds to LUMI with the new result of $\mu = 14.1$. The other marks correspond to matter-wave interferometry with molecules (green circle), atoms (purple square), neutrons (blue cross) and BECs (red plus) [93, 146, 147]. **Right:** Matter-wave interferometry can exclude certain areas in the parameter space of CSL theories. The bounds are set by assuming that a 50% reduction of the expected visibility due to a spontaneous collapse of the wavefunction can be ruled out. The blue area corresponds to the exclusion set by the former KDTLI experiment [93] and the green area corresponds to the limit set by LUMI [15]. The two points with the error bars correspond to values for the parameters λ and r_c that were proposed by Adler [169]. Both images are taken from reference [15].

in the molecular library $m = 26.777$ amu, the scaling factor as the fidelity $f = 0.93$ and the time the molecules spent in the interferometer as the coherence time $\tau = 7.5$ ms, we achieve a macroscopicity of $\mu = 14.1$. This is one order of magnitude higher than the previous interferometric macroscopicity record, set by the former KDTLI experiment [93]. Since the macroscopicity also indicates how well a non-linear theory can be excluded, we can also draw new boundaries for the CSL parameters r_c and λ as discussed in 2.2.1 (fig. 4.6). The macroscopic superposition of the oligoporphyrins can rule out new regions in the parameter space and for the first time starts to exclude the parameters proposed by Adler [169]. Although other non-interferometric tests that are looking for spontaneous heating effects [170] can exclude larger areas, matter-wave interferometry is considered a direct and more robust method in testing CSL models [171].

4.2 Anti-Helmholtz coils

LUMI was extended by a pair of anti-Helmholtz, extending the deflection measurement to particles exhibiting a permanent magnetic moment. In order to calculate the effect of the coils onto the interfering particles, the magnetic fields have to be characterized precisely. Although we can use the numerically calculated fields for our simulations, they still require a confirmation with a proper measurement of the fields inside the coils. To further use the coils for metrology measurements, we have to calibrate the magnetic system within the interferometer.

4.2.1 Characterization of the anti-Helmholtz coils

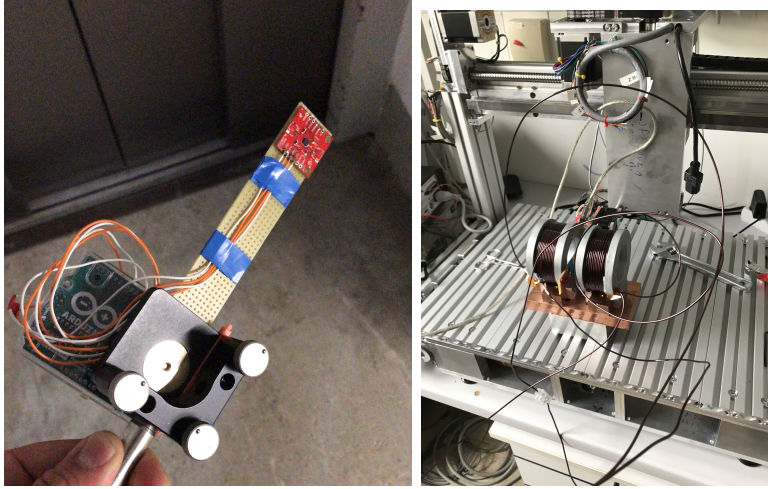


Figure 4.7: **Left:** Image of the customized magnetic field sensor assembly using an *Arduino* and magnetometer outbreak which were mounted onto a kinematic mount for round optics. **Right:** The setup that was used to measure the magnetic field inside the coils. The sensor assembly was mounted onto a custom made CNC mill, while replacing the milling head. The mill and the sensor were controlled fully automated using a *Labview* program.

We assembled a customized magnetic field sensor using an *Arduino* and a magnetometer outbreak (*Melexis MLX90393*) to characterize the magnetic fields inside the coils. The custom made assembly had the major advantage that

the sensor was small enough to fit inside the coils, while the hall probe was measuring all three field components simultaneously. The measurement was performed fully automated by using a custom built CNC milling machine ¹, where we replaced the milling head with our magnetic sensor device (fig. 4.7). The whole setup was connected and controlled via a *Labview* program and the data was analyzed and compared to our calculated fields using *Matlab* (see appendix fig. .10 - fig. .12).

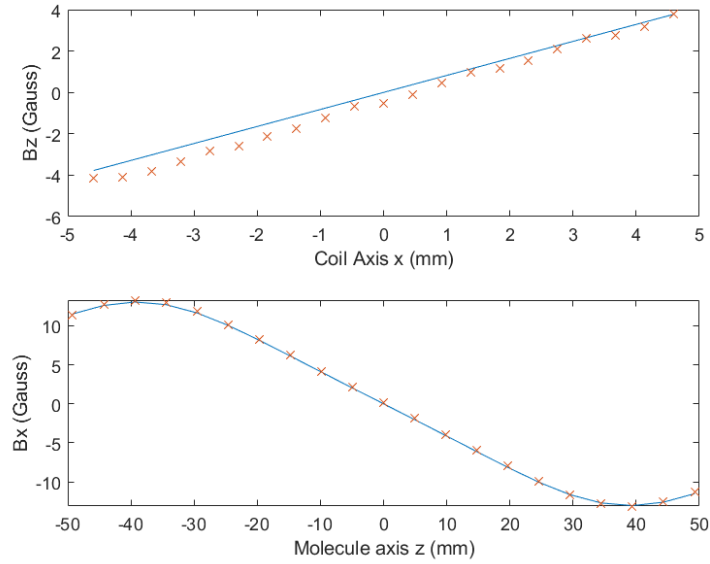


Figure 4.8: Simulated fields (solid blue line) and recorded magnetic field (red crosses) in the center of the coils. **Top** The magnetic field component B_z along the coils axis. The measured field seems to have a different slope than the expected field, which can also be observed in the field plots (see appendix fig. .10 - fig. .12). However, this can most likely be attributed to a geometrical inclination of the sensor, as the absolute field inside the coils is in perfect agreement with the calculations. **Bottom** The magnetic field component B_x along the molecular beam axis.

Initially we calculated the fields assuming ideal coils (no wire thickness) but since the wiring of the coils was done by hand ², we had to consider the dimensions of

¹Special thanks to Dr. Philipp Geyer.

²Due to the necessary thermal properties of the coils we had to use rather thick wires. This made the wrapping of the coils a demanding job and gave rise to irregularities.

the wiring (OD = 2.08 mm) and the numbers of turns per layer (15,13,12,12). Taking this into account, the fields inside the coils matched our calculations almost perfectly (fig. 4.8, fig. 4.9). The deviation that can be identified in the slope of fig. 4.8 (and also in the fields, see appendix fig. .10 - fig. .11), can be explained by a purely geometrical inclination of the sensor with respect to the coil. This assumption is also confirmed by the measurement of the absolute magnetic field inside the coils, which is again in perfect agreement with our calculations (fig. 4.9).

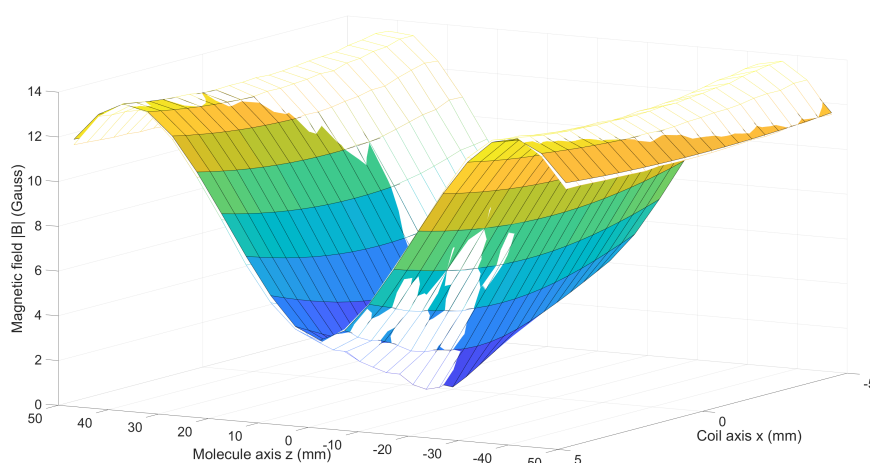


Figure 4.9: Comparison of the measured absolute magnetic field $|\vec{B}|$ inside the coils with the calculate fields. The fact that the data and the calculation match almost perfectly confirms our assumptions of a geometrical inclination of the sensors with respect to the coils as the origin of the tilted fields depicted in fig. 4.8 and fig. .10 - fig. .11 (see appendix).

4.2.2 Interferometric deflection of cesium atoms

For the calibration of the coils, we used cesium as a reference because it allows the generation of intense stable particle beams and interference of cesium was already shown at LUMI before [12]. On the other hand the ground state electronic structure of cesium has a single electron in the outer shell. Additionally, the permanent magnet moment that comes with the unpaired electron spin is well suited to calibrate the anti-Helmholtz coils.

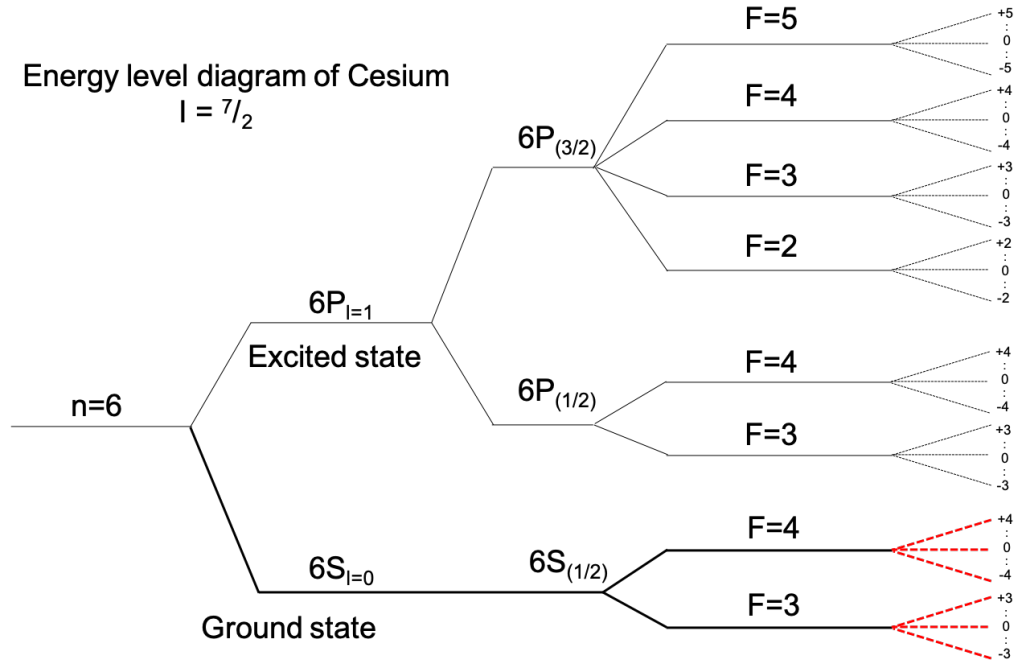


Figure 4.10: Energy level diagram of ^{133}Cs . The two hyperfine ground states $F = 3$ and $F = 4$ are split into 7 ($m_{f,F=3} = -3, ..0, ..3$) and 9 ($m_{f,F=4} = -4, ..0, ..4$) magnetic states.

Electronic structure To understand how the magnetic field of the coils acts on the interfering particle, we have to understand the magnetic properties of the atom. Considering the unpaired electron in the outer shell of the alkali metal, one might expect possible permanent magnetic moments of $\pm\mu_B$, as it was demonstrated in the example simulation (fig. 3.10). Given the extreme

sensitivity of LUMI, however, one has to also consider the hyperfine structure with all its magnetic projections. Cesium is a monotopic element (^{133}Cs) holding a nuclear spin of $I = 7/2$ and an electron configuration of $[\text{Xe}] 6s^1$. The interaction of the nuclear spin with the electron cloud leads to a hyperfine structure with two hyperfine levels $F = 3$ and $F = 4$ in the ground state (fig. 4.10). Additionally we have to take into account the perturbation of the energy levels due to the external magnetic field which is present inside the coils. Since the magnetic field strengths of the coils are below 100 Gauss for the currents we use, we are operating in the low-field regime (fig. 4.11). This means that the Zeeman energy splitting depends linearly on the magnetic field with $\Delta E_{|F,m_f\rangle} = \mu_B g_f m_f B_z$, with μ_B the Bohr magneton, g_f the total spin g-factor and m_f the magnetic quantum number [172]. The hyperfine states of the ground states are separated into a total of 16 possible magnetic states (fig. 4.10).

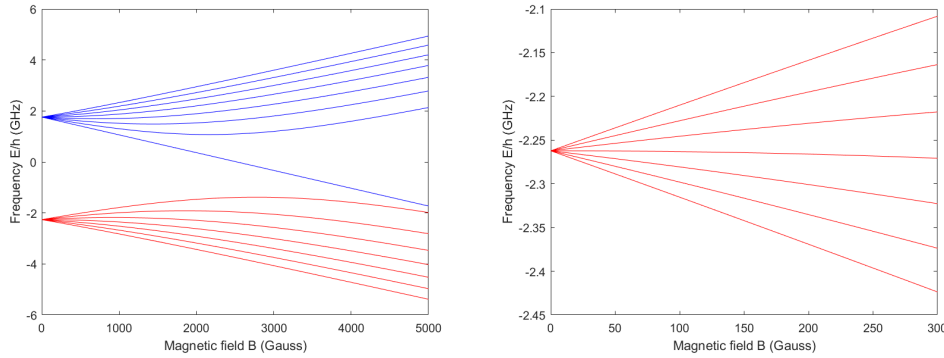


Figure 4.11: Energy splitting of the ^{133}Cs hyperfine levels in a magnetic field in dependence of an external magnetic field. The magnetic fields of the anti-Helmholtz coils operate in the low field regime (zoomed in box), where the splitting is linear with the magnetic field.

We calculate the magnetic moment μ of the atom using the magnetic quantum number m_f and the total angular momentum F , which depends on the coupling of the spin angular momentum S , the orbital angular momentum L , with $J = L + S$ and the nuclear spin momentum I

$$\mu = g_F m_F \mu_B, \quad (4.2.1)$$

with the magnetic numbers $m_{f,F=3} = -3, ..0, ..3$ and $m_{f,F=4} = -4, ..0, ..4$ and the g -factor which can be calculated by

$$g_F = g_J \frac{F(F+1) - I(I+1) + J(J+1)}{2F(F+1)} + g_I \frac{F(F+1) + I(I+1) - J(J+1)}{2F(F+1)}. \quad (4.2.2)$$

The contribution of the nuclear momentum is in general much smaller and can be neglected for our further calculations [172]. Using the simplified formula we calculate the g -factors $g_3 = -0.125$ and $g_4 = 0.125$. Taking all resulting 16 magnetic moments into account we can calculate the shifts according to eq.3.2.7 and derive the reduction of contrast due to the different accumulated phase shifts inside the coils.

Experimental protocol The handling with alkali elements always requires some special care. Owing to the high reactivity of cesium we had to fill the oven in a glove box with an inert gas environment. The most difficult part of the filling process was to bring the oven back to the source chamber of LUMI without exposing the sample to air. However, with a little skill we managed to conduct our experiments without any special occurrences. Once the oven was back at LUMI and under vacuum the experiment was handled like with any other atom or molecule.

During the experiment we have selected the velocities of the thermal beam gravitationally by using horizontal slits. Given a Gaussian fit, the mean velocity of the atom beam was about (519 ± 117) m/s and aligned to pass through the smaller slit (0.5×0.5 mm², see fig. 3.9). After traversing the interferometer, which was operated in the TLI scheme (MMM, $d=266$ nm), the atoms were ionized with a rhenium hot wire, mass-filtered and detected. Before starting the actual deflection measurements we recorded several reference interference scans with the coils turned off. Although the cesium oven provided high count rates

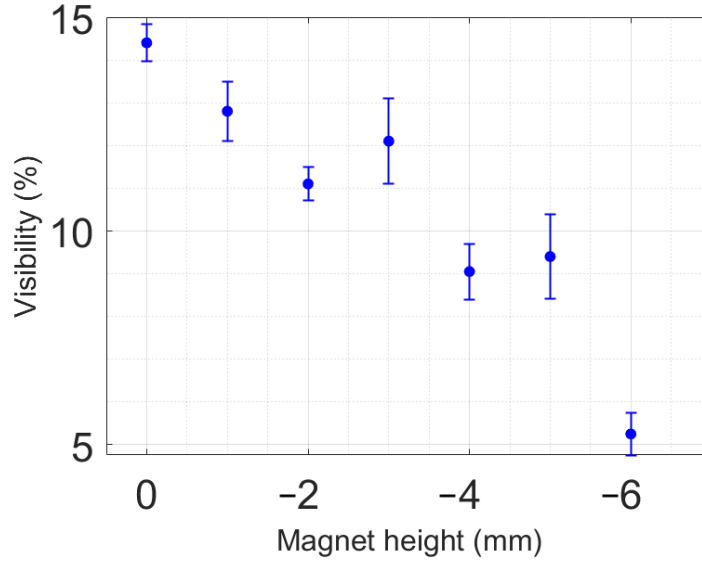


Figure 4.12: Measured visibility of non-deflected interference scans of cesium atoms as a function of the height of the permanent magnet. The height of the permanent magnet is referenced with 0 corresponding to 'fully withdrawn'. We can see that the magnet, which is mounted between the first and the second grating (fig. 3.8) reduces the contrast of the interference. The curve indicates that the stray fields have an effect on the permanent magnetic moment of the atoms even when the magnet is fully withdrawn from the molecular beam, while the effect gets stronger as we move the magnet closer to the beam.

and an excellent signal-to-noise ratio, we failed to achieve the full theoretically expected contrast. We attribute this to the permanent magnet, which was still installed during these measurements. The remanent fields of the Halbach magnet affected the cesium atoms even with the magnet fully withdrawn (fig. 4.12).

For the actual deflection measurements we tuned the current through the coils in steps of 0.2 A up to 16 A, while recording two interference scans for each step. During the measurement it was indispensable to always monitor the temperature of the coils as well as the pressure inside the interferometer chamber. Particularly for higher currents the assembly becomes rather hot which in turn leads to a significant outgassing of the coils.

Results According to our calculations we should see a complete loss of the contrast at already low coil currents of about 1 A and a first revival at around 3 A. For higher currents, the contrast should finally asymptote to a value corresponding to $1/8$ of the initial contrast.

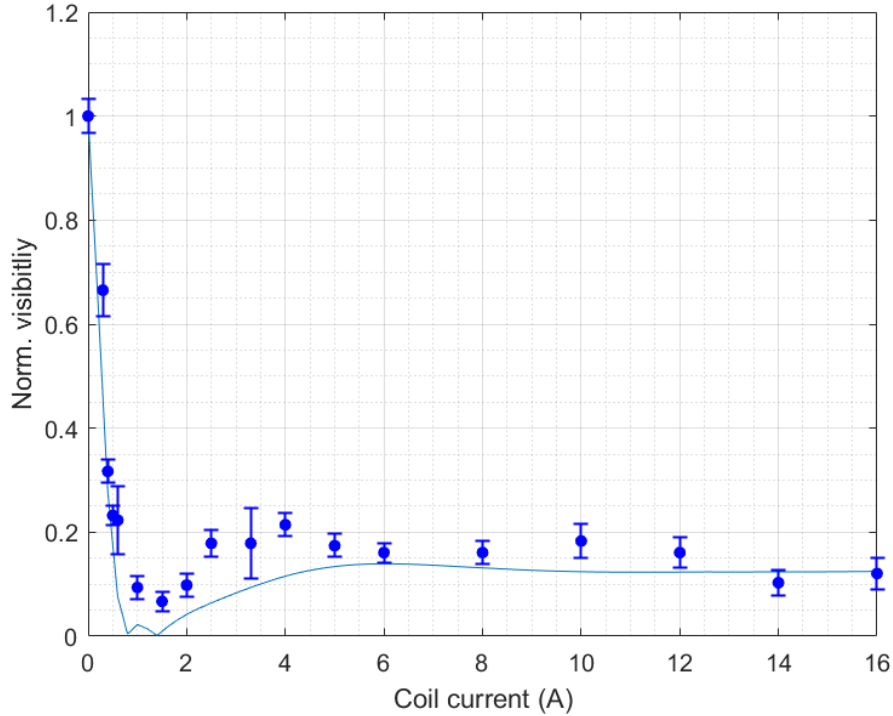


Figure 4.13: Visibility of the deflected cesium atoms as a function of the current through the coils. The data is in good agreement with our simulation and asymptotes at a value of $1/8$ of the initial visibility. A drop at higher currents can be explained by collisional decoherence. Due to the higher temperatures of the coils the pressure started to rise significantly. Collisions between the cesium atoms and the outgassing particles can be attributed to the reduction in contrast arising at higher currents.

The remaining $1/8$ of the initial visibility is owed to the fact, that two of the 16 electronic states come with a vanishing magnetic moment due to $m_f = 0$. These two states are not shifted at all in the coils and therefore experience no loss of contrast due to velocity averaging. The recorded experimental data fits quite well with our simulation (fig. 4.13). The drop of the contrast and even

the revival can be identified at the predicted coil currents. The repeated drop in contrast, which can be observed at higher currents could be explained by collisional decoherence. Due to the higher currents the temperatures start to rise and the numbers of collision with particles from the outgassing coils start to reduce the contrast.

Conclusion The implementation of the anti-Helmholtz coils extended the metrological capabilities of LUMI to particles exhibiting a permanent magnetic moment. The deflection measurements of cesium demonstrated the high sensitivity of the interferometer, as it is required to take all hyperfine states and the corresponding Zeeman effect into account to fully explain the shifted interference fringes. In order to further confirm the calibration of the magnetic system, we will have to repeat the deflection experiment with a different particle. For a proper calibration it is advisable to remove the permanent magnet before the measurements are repeated.

Chapter 5

Outlook

We have successfully shown that LUMI is compatible with high mass interference experiments and can serve as a sensitive tool for advanced metrological measurements. That is why it is promising to keep the course and to pursue further experiments at LUMI. In this chapter I will give a brief overview of some of the planned short and medium-term steps that will extend the capabilities of LUMI even further. These are at one hand applied deflection measurements with the anti-Helmholtz coils and on the other hand the upgrade of LUMI to an all-optical interferometer, pathing the way for metal cluster interference with masses up to 500.000 amu [6].

5.1 Metrology with the anti-Helmholtz coils

The anti-Helmholtz coils allow us to measure permanent magnetic moments interferometrically, as has been shown in this thesis for cesium. In order to confirm our calibration, another measurement with rubidium will be taken. After calibrating the magnetic system with atoms, we can use the coils for deflection measurements with a large variety of molecules and metal clusters. The precise control over the magnetic field and the optical access of the coils allow us to set up photo-switching experiments. This can be done by transversely intersecting a laser beam with the molecular beam inside the coils (fig. 5.1). The light in-

teracts with the molecules and can induce a change of their electronic structure and subsequently of their magnetic properties. One possible application is the measurement of the photo-induced triplet state excitation of fullerenes. The triplet excitation of C_{60} can efficiently be carried out with our existing laser systems at wavelengths ranging from UV to green light [173]. The absorption of the photons excite the fullerenes into the lowest excited singlet state which then internally decays into the long-lived triplet states. Since the absorption cross-section is well known we can gain access to the lifetime of the triplet states by measuring the deflection as a function of the laser power.

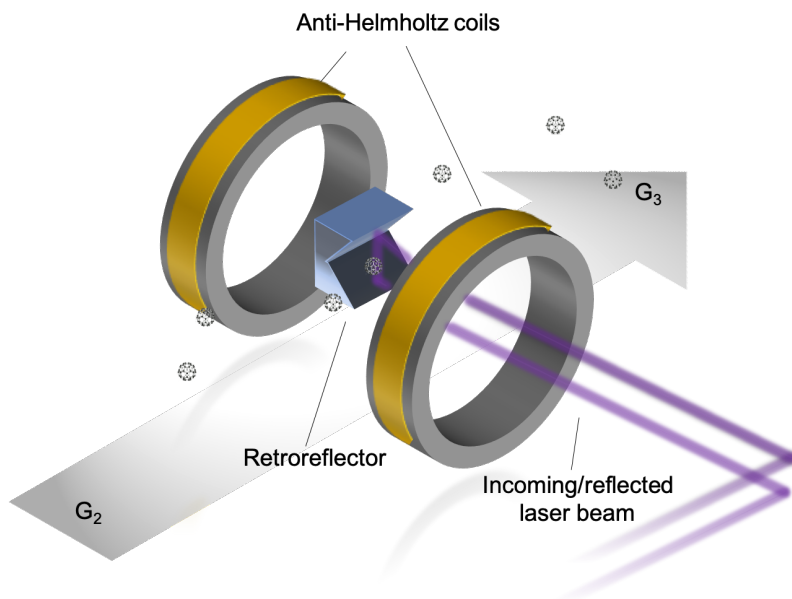


Figure 5.1: Schematic diagram of the photo-switching experiment. The optical access of the coils allows us to introduce a laser beam, intersecting with the passing molecular beam. To avoid dephasing due to transverse recoil kicks from the photon absorption, the laser beam crosses the molecules vertically. This is realized by placing a retroreflector mirror inside the coils. When the absorption cross-section of the molecule is known we can measure the lifetime of photo-induced triplet states by recording the deflection of the beam as a function of laser power.

5.2 Metal cluster interference

LUMI succeeded in high-mass interferometry by showing superposition of molecules with masses up to 28.000 [15]. However, the mass-velocity product was already situated at the border of what is possible with the current configuration of LUMI (fig. 5.2). In order to push the limits to even higher masses we are going to upgrade the interferometer by realizing an all-optical interferometer and combining it with a cluster aggregation source.

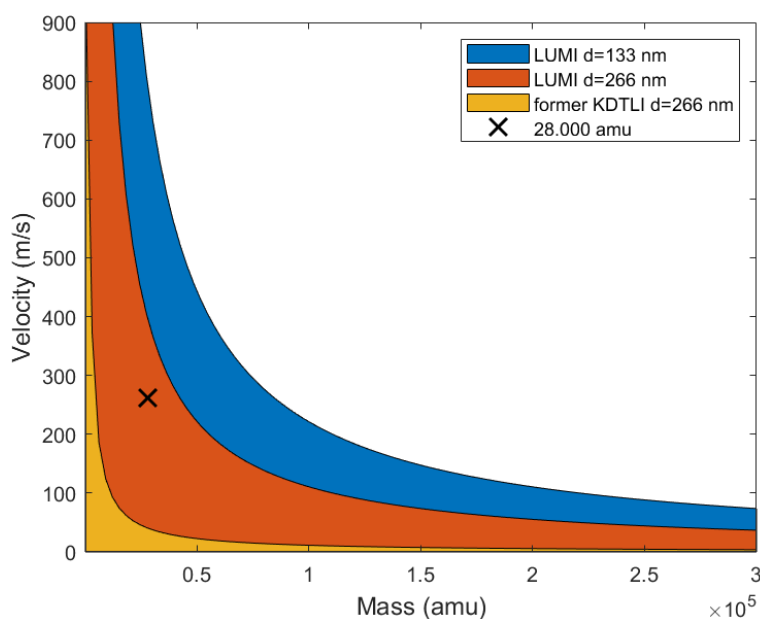


Figure 5.2: Compatible mass - velocity products for LUMI consisting of three optical depletion gratings with a period of 133 nm (blue), for the current setup of LUMI consisting of two material and one optical phase grating with a period of 266 nm (red) and the former KDTLI experiment with the same interferometer scheme as the current LUMI but a reduced baseline length of 0.1 m (orange). The black cross, situated close to the border of the actual setup, represents the high-mass interference of the oligoporphyrins at LUMI [15].

Optical depletion gratings

Whenever the photon energy exceeds the ionization energy of a cluster, a standing light wave can in principle be used as an optical depletion grating. In this case, absorption of a single photon suffices to ionize the cluster independently from resonances. The optical grating depletes the incident cluster beam such that the clusters are ionized in the antinodes. Taking a look at the periodic table, we can identify over 30 metals with work functions which are at least 0.3 eV below the energy of a 266 nm photon (4.66 eV). The lower threshold is needed because the work functions are usually defined for bulk material and the ionization energy of clusters is usually higher. The low electron affinities of metal clusters also allow to introduce neutralization depletion gratings, whereby singly charged ions can be neutralized by single photons.

The ionization energy approaches the work function as the clusters grow in size [174]. Assuming 0.3 eV should cover most of the metals for cluster sizes exceeding 100 atoms [5]. The work function of cesium is only $W = 2.1$ eV and green photons (532 nm) come with sufficient energy (2.33 eV) to ensure single-photon ionization. Since high continuous laser powers are available for this wavelength and the optics are already integrated in LUMI, it is worth thinking about an intermediate step before upgrading to three UV-gratings. For both schemes, either UV or green optical gratings, it is required that an average of at least one photon must be absorbed in the antinodes while the molecules pass the standing light wave to act as a coherent beam splitter. According to the number of photons absorbed in a standing light wave (eq.2.1.13) we notice that already low laser power suffices to fulfill this condition.

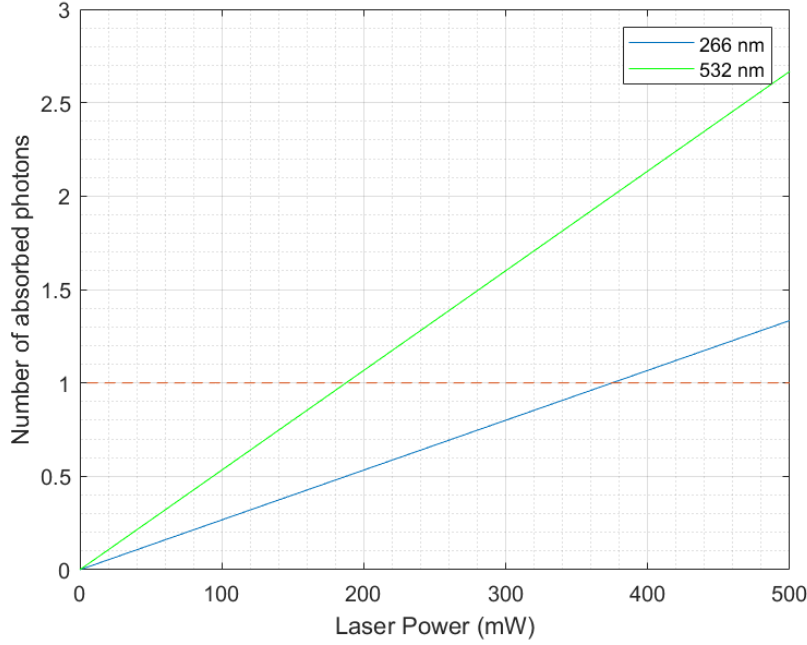


Figure 5.3: Number of absorbed photons for a given laser power and photon wavelength $\lambda = 266$ nm (blue) and $\lambda = 532$ nm, assuming a TEM_{00} laser profile with a beam waist of $w_y = 1$ mm, a particle velocity of $v = 150$ m/s and an absorption cross section of $\sigma_{abs} = 10^{-15}$ cm², which is valid for most of the metal clusters consisting of more than 100 atoms [175].

We have designed the optical setup such that LUMI 2.0 can be operated in both schemes. The 20 W of green light from a *Coherent Verdi V20* laser system can serve as a pump laser for a second harmonic generator (SHG) or directly be used as a green optical grating. Two additional photo-detachment stages are intended as a neutralization mechanism before entering the interferometer and as an ionization mechanism before the detection takes place.

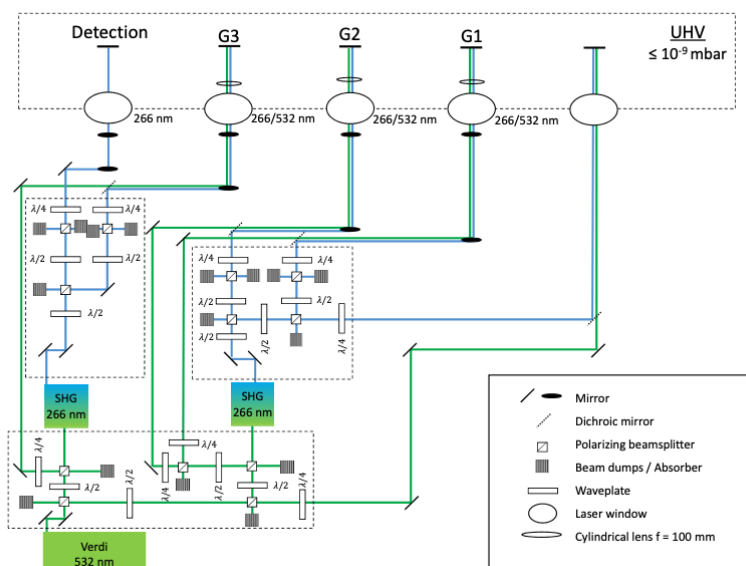


Figure 5.4: Schematic diagram of the optical setup of LUMI 2.0. This setup allows the operation of three optical gratings and two additional photo-detachment stages with either green light (532 nm), blue light (266 nm) or a mixture of both. The initial 20 W green light from the *Coherent Verdi V20* laser system can be distributed individually between the gratings or the photo-detachment stages. Two (*Newport Wavetrain SHG*) can be pumped with up to 8 W of green light to generate UV-light (266 nm) with an output power of approximately 1 W. The arrangement of double-coated optics allow us to use both schemes without having to replace the optics and distribute the laser powers wherever needed.

Cluster aggregation source

Cluster aggregation sources produce a beam of clusters by vaporizing material into a flow of cold inert gas. Due to the low temperatures of the gas the vapor approaches supersaturation and initializes the cluster-formation [111]. This technique can in principle be combined with all the fore-mentioned source methods as a vaporization mechanism. Another well-working technique of cluster beam generation is magnetron sputtering. A metal surface is bombarded with highly energetic ions, sputtering material from a target. The combination of the magnetron sputtering source together with the gas aggregation method

can be used to produce clusters from all solid elements. Since the highly energetic ions tend to sputter dimers, trimers and larger pieces [176], it is on the contrary to the vaporization-based aggregation sources, not dependent on the condensation process of single atoms, which is beneficial in the formation of large clusters [111].

Magnetron sputtering The principle of a magnetron sputtering source is well known and widely used in physical chemistry and material sciences as a nano deposition source. The target, which is made out of the desired material, is connected to a high voltage supply and takes on the role of the cathode.

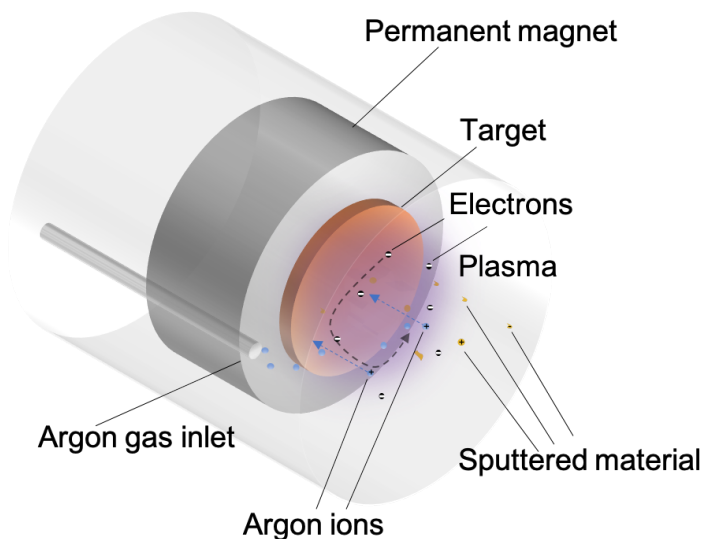


Figure 5.5: Schematic diagram of magnetron sputtering head. The material target is mounted in front of a permanent magnet enclosed in a cylinder and inert gas flows into the surrounding cell. A plasma is ignited by applying high voltages between an electrode and the target plate. While the magnetic field forces the electrons to follow a helical trajectory, the more massive ions are accelerated almost undisturbed towards the target, where they sputter material on impact. The sputtered material is then swept into a condensation cell which is filled with argon or helium to form clusters. Additional cooling down to 77 K is achieved by flooding the double-walled tube with liquid nitrogen. After trespassing the condensation cell, the cluster beam exits through an orifice or an adjustable iris into the vacuum chamber.

For sufficient gas loads and voltages, the electric field ignites a plasma in front of the target. While the electrons are repelled, the positively charged ions are accelerated towards the target. A permanent magnet placed behind the target forces the electrons on helical trajectories, increasing the residence time. The mass-dependent Larmor radius $r_L = \sqrt{2m/U|q|B}$ of the trajectories reveals why the inert-gas ions remain mostly unaffected on their way to the target. At the same time it is desirable to choose the parameters such that a high electron density in front of the target is achieved. This in turn increases the numbers of ions and therefore also the sputtering efficiency. Due to the interaction of the sputtered material with the plasma the cluster beam includes neutrals, singly negative and singly positive charged ions. To further enhance the clustering, the molecules are then cooled by collisions with cryogenic cooled gas atoms inside an aggregation cell. The cluster sizes are typically log-normal distributed while the properties of the distribution depend strongly on the aggregation time. [177]

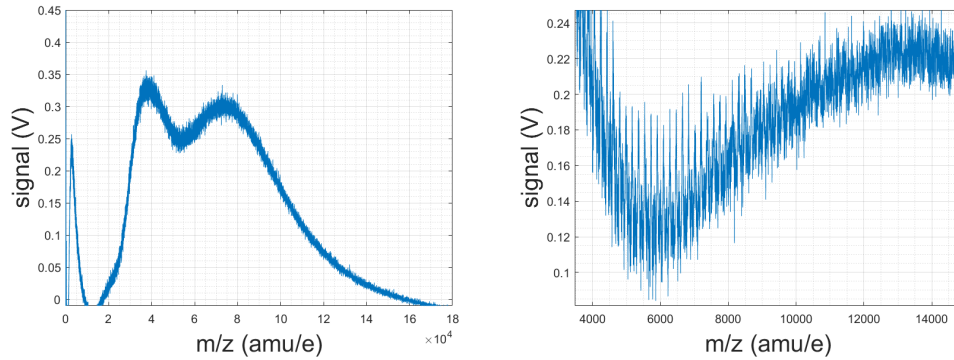


Figure 5.6: Time-of-flight mass spectra of hafnium clusters produce with a magnetron sputtering source **Left** Hafnium clusters consisting of up to 1100 atoms. **Right** A closer look at smaller masses reveals the different cluster sizes on an atomic level ($m = 178.5$ amu).

Hafnium has a work function of 3.9 eV and atomic mass of 178.5 amu. It is therefore promising candidate for UV-depletion gratings and high-mass metal cluster interference. For our first tests we have used a cluster aggregation source with magnetron sputtering, provided by B.v. Issendorff from the University of Freiburg. We were able to produce hafnium clusters with masses up to 200.000

amu, while being able to resolve small clusters on an atomic scale (fig. 5.6). However, further tests to characterize the source and to better control the process of cluster formation have to be performed before we can implement the source in LUMI.

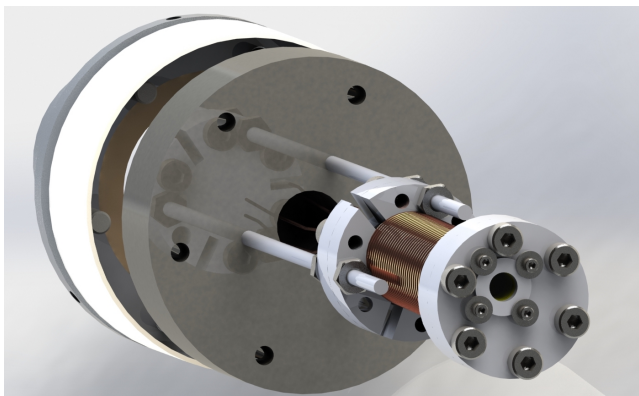


Figure 5.7: Many designs for thermal beam sources have been proposed and used to produce beams of alkali atoms and all of them have in common that the metals are kept isolated from environmental air during the filling process and until the actual application of the source. We have designed this oven to be used within our cluster aggregation source, replacing the sputtering head with the oven assembly. It is in principle an effusive cell with the main difference that the orifice is large ($d = 3$ mm) and placed off-center to avoid clogging and spilling over when the cesium becomes fluid. The orifice can also be tightly sealed with a paraffin film using a screwed-on metal ring with a lip. This was helpful to avoid exposure of the cesium with air during the installation in the vacuum chamber. The paraffin film could then simply be melted away inside the vacuum chamber.

Alkali metals Due to their low ionization energies, alkali metals are particularly suited for photo depletion gratings. Although the atoms can be easily volatilized with thermal ovens, their low melting points exclude them for being used in sputtering sources. Cesium, for example, has a bulk work function of 1.95 eV [178] and a single green photon (532 nm) has sufficient energy to ionize the clusters. Since the second grating of the current LUMI setup is already established by a standing light wave from a doubled Nd:YAG laser (532 nm), interference experiments of alkali clusters could be an intermediate step before swapping the entire interferometric setup to UV-gratings. For our first tests

we have therefore designed an oven for alkali metals (fig. 5.7) which was compatible with the aggregation source. The oven was installed instead of the sputtering head, while every other part of the aggregation source remained as it was.

Using this assembly we were able to produce cesium clusters consisting of up to 1500 atoms (fig. 5.8). However, the cluster beam was contaminated with paraffin and the signal only lasted for about 15 minutes.

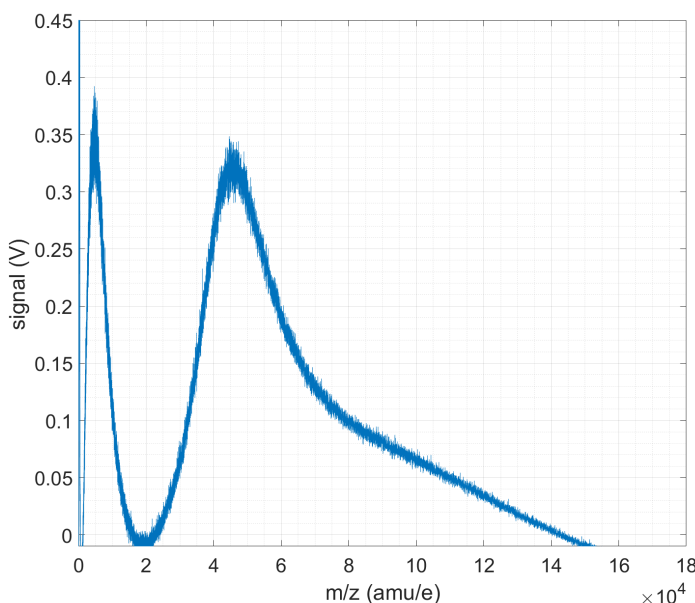


Figure 5.8: Time-of-flight mass spectrum of cesium clusters consisting of up to 1500 atoms. The spectrum is probably contaminated with paraffin from the oven sealing. The clusters are produced by thermally vaporizing within a cryogenic cooled aggregation cell filled with argon. A specially designed oven (fig. 5.7) was therefore mounted inside an existing cluster aggregation source, replacing the original magnetron sputtering head. The signal lasted for about 15 minutes and we were not able to resolve single cesium peaks. The peak at $\sim 1.85 \times 10^5$ amu originates from electronic noise only.

Also the chamber was coated with a film of paraffin which is not beneficial for the vacuum system. Even though we can get rid of the paraffin by adapting the oven design, we would still require the signal time to be extended many

times over. The lack of robust sources will make the implementation of an intermediate LUMI 1.5 with three green optical gratings challenging.

Experimental setup

In addition to the existing parts, the upgraded setup of LUMI 2.0 will include a cluster aggregation source, an ion manipulation including rf-guiding, mass-selection and buffer gas cooling and three optical UV-gratings with a period of $d = 133$ nm.

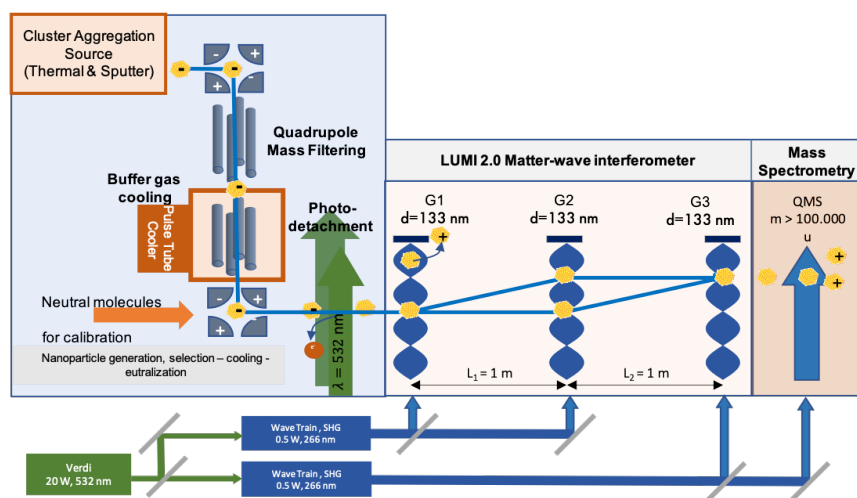


Figure 5.9: Schematic diagram of the experimental setup of LUMI 2.0 with three optical depletion gratings. The cluster aggregation source with a thermal oven or magnetron sputtering, produces clusters with a charge distribution of approximately 1/3 neutral, 1/3 positive and 1/3 negative, with masses beyond 10^6 amu. While the neutral clusters will be filtered out by the first bender, the singly charged ions are guided to the first QMS and to the buffer gas cooling cell. The mass-selected, internally and transversely cold ions are then again steered by a bender into the interferometer. The second bender will be situated in the current source chamber of LUMI and allows us to attach any other molecular beam source available for LUMI. This will be particularly useful for alignment procedures. After passing the second bender, the ions are neutralized by a photo-detachment laser or by the first depletion grating. In both cases only the neutral particles are kept within the beam. The second grating will then deplete the beam by ionization. The third grating will again act as an ionization grating. The resulting ions can either be used directly for the detection or removed from the beam. In the latter case the neutral particles would have to be ionized by an additional stage before they can be detected. Adapted image provided by Prof. Markus Arndt.

We can take advantage of the fact that the cluster source produces singly charged particles and manipulate the ions using ion optics. A combination of benders and quadrupoles will allow us to mass-select, cool and guide the ions to the interferometer. A photo-detachment stage can either be implemented in front of the interferometer or the first grating can be used as neutralization depletion grating, to ensure neutral particles throughout the coherent part of the interferometer. The second grating will then act as a phase grating or a depletion grating, removing the particles that are ionized from the beam. The third grating will again be an ionizing grating. The ions can then be directly used for the detection or again removed from the beam. With the latter, an additional ionizing stage for the detection has to be included. This can either be photo-ionization, electron impact ionization or Langmuir-Taylor surface ionization.

Conclusion

The upgrade to an all-optical scheme with the combination of a cluster aggregation source will make LUMI fit for future macroscopic interference with metal clusters up to 300,000 amu. Aggregation sources with magnetron sputtering are particularly promising for high-mass metal cluster interference. The sources are capable of producing intense beams of neutral and charged clusters with masses beyond 10^6 amu. The low work function of some metals allows us to introduce photo-depletion gratings with available laser systems. Considering a 266 nm laser beam for the optical gratings, we can identify more than 30 elements whose ionization energy is at least 0.3 eV below the photon energy. The resulting 133 nm period of the standing light waves will make LUMI compatible with de Broglie wavelengths down to 20 fm. Due to their low work function, alkali metals are particularly tempting candidates for optical depletion gratings. For cesium, even the current optical grating ($\lambda_L = 532$ nm) suffices to ionize the atoms. However, preliminary tests showed that the high reactivity and the low boiling points make it difficult to implement an intermediate version of LUMI 2.0. The upgrade version will also allow us to extend the advanced

metrological sensitivity to a new material class. In this way we can investigate magnetic, electrical or optical properties while they undergo a transition from semi-condensed matter to condensed matter. Until then, many tests have to be done outside of the interferometer. The cluster source has to be fully characterized together with the metals we plan to use. We have to study the clusters, test the depletion efficiency and detection mechanism. Furthermore, we have to set up and test the whole test chambers for the beam preparation and ion manipulation. The optical setup for the gratings has already been designed, but still needs to be set up, tested and installed in the interferometer. Some of the listed work has already started but it will take some effort and time before we will finally see interference of metal clusters on LUMI 2.0

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Appendices

.1 Magnetic field measurements of the anti-Helmholtz coils

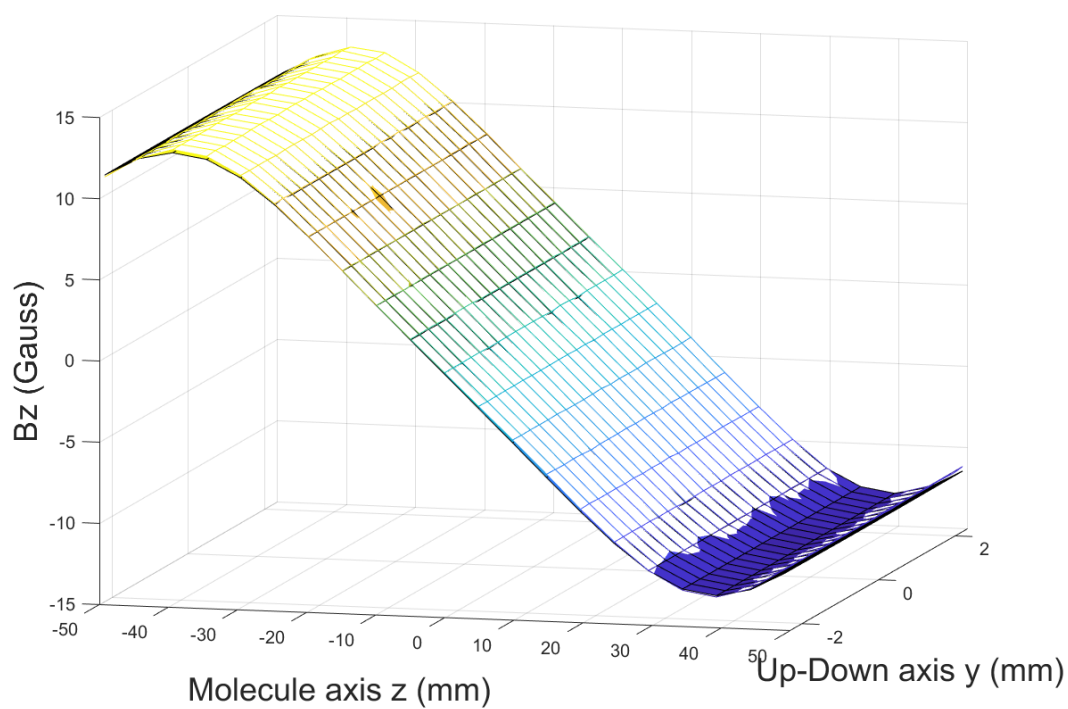


Figure .10: Measured magnetic field $B_z(x = 0, y, z)$ (meshed area) inside the coils in perfect agreement with the simulated fields (solid area).

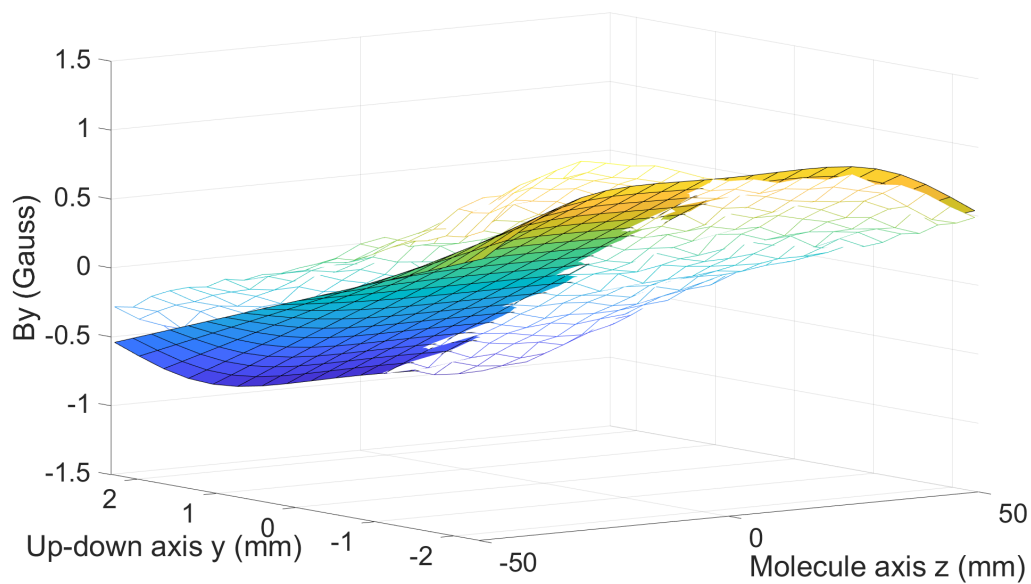


Figure .11: Measured magnetic field (grid) compared to the simulated fields (solid). **Left:** $B_y(x = 0, y, z)$ **Right:** $B_z(x, y = 0, z)$. Both measurements show a slightly tilted field compared to the calculations. This can be explained by purely geometric inclination of the magnetic sensor w.r.t. the coil.

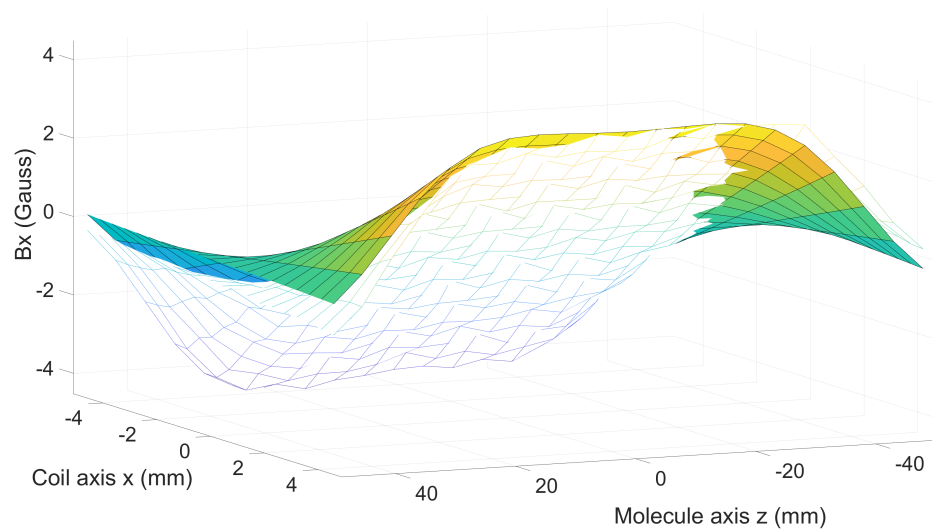


Figure .12: Measured magnetic field (grid) compared to the simulated fields (solid). **Left:** $B_y(x = 0, y, z)$ **Right:** $B_z(x, y = 0, z)$. Both measurements show a slightly tilted field compared to the calculations. This can be explained by purely geometric inclination of the magnetic sensor w.r.t. the coil.

.2 Series of interference scans: 28.000 amu

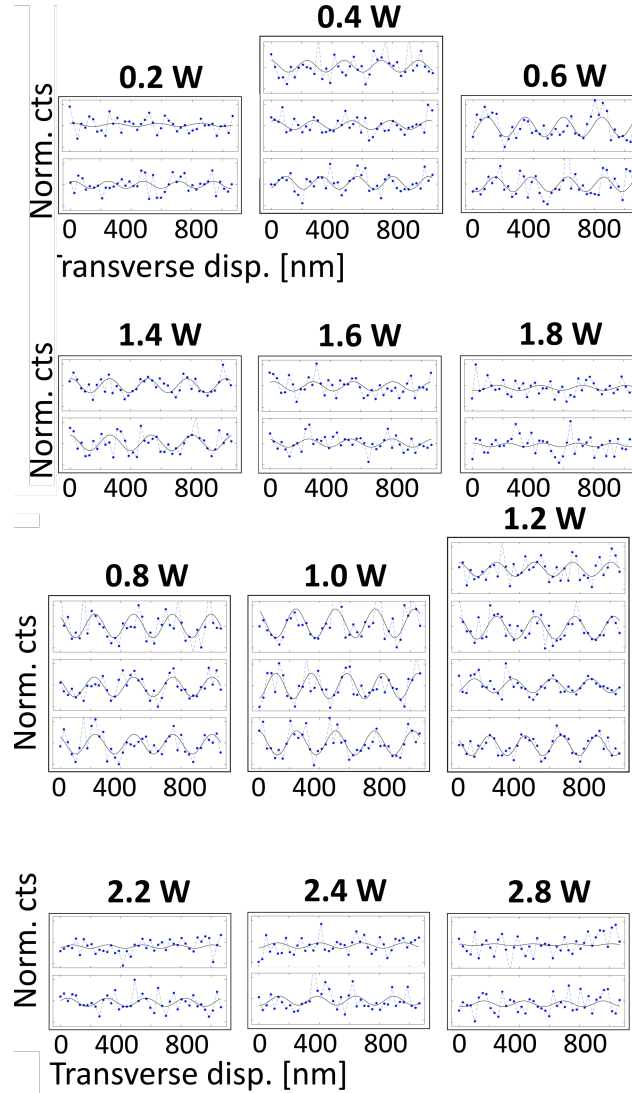


Figure .13: Series of the interference scans with increasing laser power of the optical grating G_2 . The maximum contrast with $V = 30\%$ was achieved at $P = 1$ W. This is in perfect agreement with the theoretical simulations for the quantum regime. Adapted image taken from reference [5].