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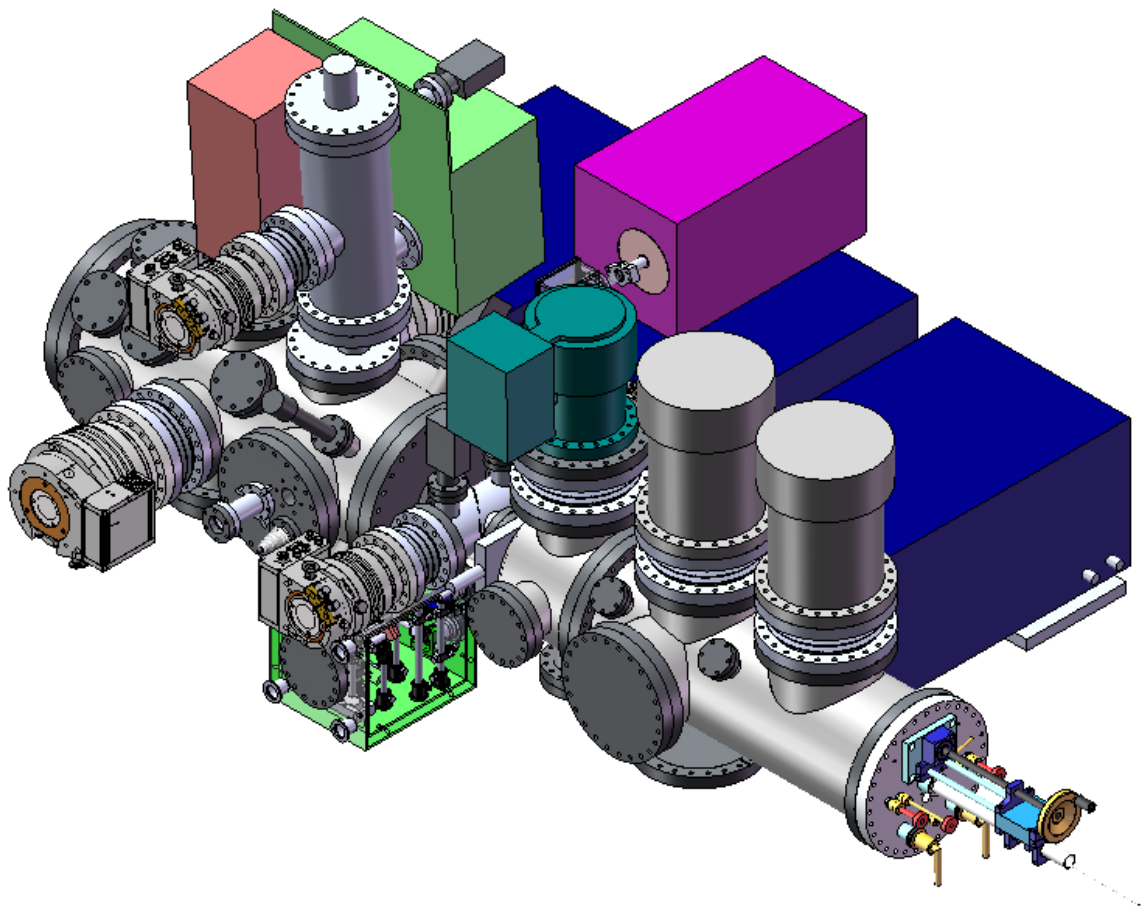
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Optical Time-domain Interferometer for Matter-waves (OTIMA)

“I, the machine, show you the world as only I can see it.”¹



¹ This quotation is known in the context of the rise of cinematography, but I find it describes the situation in modern experimental physics quite well also. Nowadays most experimental physicists don't see the results of the experiments they do with their own eyes, but only through the sensors, detectors and algorithms of their intricate machinery.

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1. Zusammenfassung

Die Quantenmechanik ist eine der meistgeprüften Theorien in der Physik, jedoch sind die auf der mikroskopischen Skala beobachtbaren Quanten-Phänomene scheinbar unvereinbar mit dem Verhalten der makroskopischen, klassischen Welt. Ob dieser Übergang vom quantenmechanischen zum klassischen Verhalten nur scheinbar oder real ist, ist bis heute eine offene Frage. Während meiner Dissertation habe ich in Zusammenarbeit mit meiner Kollegin und meinen Kollegen ein Talbot-Lau Interferometer aufgebaut und betrieben, welches erlaubt, Quanteninterferenzen massiver, komplexer Teilchen zu untersuchen. Damit sollen sich in zukünftigen Generationen des Experiments auch neue Hypothesen des Übergangs zur klassischen Welt prüfen lassen. Im Rahmen meiner Arbeit habe ich das Interferometer mittels CAD geplant und am Aufbau maßgeblich mitgewirkt. Ich habe das Software System MOPS (*Molecular Optics Programming System*) zur Datenerfassung und Versuchsablaufsteuerung entworfen und implementiert. Ferner habe ich verschiedene Molekularstrahlquellen entworfen und getestet, um für das Experiment interessante Teilchen ladungsneutral in die Gasphase zu bringen, so dass ihre Geschwindigkeit und Intensität den praktischen Anforderungen des Interferometers genügen. Unser *Optical Time-domain Interferometer for Matter-waves* (OTIMA) besteht aus drei in sich selbst reflektierten ~ 7 ns kurzen Excimer-Laserpulsen mit einer Wellenlänge von 157.6 nm. Sie erzeugen optische absorptive (*photodepletion*) Gitter mit einer Periode von 78.8 nm. Da der Aufbau ohne materielle Beugungselemente auskommt, gibt es keine dispersiven van-der-Waals Kräfte, die das Interferenzmuster auswaschen würden. Auch ist die Spaltbreite jedes Gitters durch die Laserenergie variabel einstellbar. Wir erwarten somit einen hohen Interferenzkontrast selbst für Teilchen hoher Masse. Unter realistischen Bedingungen auf der Erde sollte dieser Interferometertypus mit Teilchen mit bis zu 10^6 amu betrieben werden können. Im Laufe unserer Experimente konnten wir bisher erfolgreich die Interferenz molekularer Cluster mit bis zu 2300 amu beobachten und wir konnten zeigen, dass nicht nur die Einphotonenionisation sondern auch die Einphotonenfragmentation von Teilchenclustern als Mechanismus genutzt werden kann, um Teilchen aus dem Strahl zu entfernen. Dadurch konnten wir eine neue Klasse von schwach gebundenen Teilchen für Materiewellenexperimente und neue Methoden für Biomoleküle erschließen.

2. Abstract

Quantum Mechanics is one of the most thoroughly tested theories in physics; however the quantum phenomena that appear on the microscopic scale are incompatible with the behavior of the macroscopic world. Whether the transition between quantum and classical behavior is virtual or real is still an open question. During my thesis I have built, together with my colleagues, a Talbot-Lau interferometer with light gratings that is capable of handling very large and complex particles. With this device it will be possible to test some of the hypotheses that postulate mechanisms for the quantum to classic transition. During my thesis I have designed the experimental setup using CAD and we assembled the apparatus. I have designed and implemented the data acquisition and experiment control software system MOPS (*Molecular Optics Programming System*). Furthermore, I have implemented and tested various particle sources for the experiment to bring neutral particles into the gas phase at a velocity and with a beam flux that meets the requirements of the experiment. The *Optical Time-domain Interferometer for Matter-waves* (OTIMA) is made up of 3 retro-reflected, ~ 7 ns short excimer laser pulses with a wavelength of 157.6 nm, i.e. a grating period of 78.8 nm. The purely optical and pulsed diffraction elements avoid all dispersive interactions that would reduce the interference contrast. Therefore, we expect a high fringe contrast even for large particles; under realistic conditions on earth this type of interferometer is conceptually capable of exploring the wave-particle duality with particles up to 10^6 amu or even beyond. During my PhD thesis we successfully showed interference for single-photon ionizable molecular clusters up to 2300 amu. Furthermore, we have demonstrated that single-photon fragmentation gratings enable interference experiments with a new class of weakly bound particles and provided interesting perspectives for biomolecules.

3. Introduction

The predictions of quantum mechanics, while astonishingly accurate, seem to contradict our everyday perception of macroscopic reality. In our experience objects have defined properties like their coordinates or velocities. In quantum mechanics such properties become uncertain [1] and particles may behave like waves [2, 3]. This was discovered by Louis de Broglie who postulated in 1923 that similar to massless photons ($m = 0$), all moving particles should behave like waves and he attributed them a wavelength:

$$\lambda = \frac{h}{m v}$$

where m is the particles mass, v its velocity and $h = 6.62 \times 10^{-34}$ Js is Planck's constant [4]. Shortly thereafter, in 1927, this daring hypothesis was confirmed by Davisson and Germer [5, 6] when they successfully observed the diffraction of electrons at a nickel crystal. Later, in 1929 Estermann and Stern [7] showed diffraction of a helium beam at a LiF crystal. The wave hypothesis laid ground to quantum physics as we know it today. The obvious question then is why we don't see quantum effects in our everyday life, if there is a fundamental limit to the quantum description of our world. Is it only due to decoherence that the world appears classical? Over several decades theoreticians developed modifications [8, 9] to quantum mechanics that all share the emergence or introduction of a non-linearity in the otherwise unitary and linear theory. One way to test such hypotheses is to probe the mass limits of quantum mechanics, trying to observe the quantum to classic transition in an experiment. A straightforward approach is to do interference experiments with particles of increasing size and complexity.

The first interference experiments with truly complex particles were done by Arndt and Zeilinger in 1999. They successfully diffracted C_{60} fullerenes, with a mass of 720 amu [10]. Diffraction at nanomechanical masks has recently been done with particles up to 1,298 amu [11]. In these setups particles have been emitted by a thermal source and collimated using slits to obtain a coherent beam. This was then diffracted at a nanofabricated grating and the emerging far-field diffraction pattern was recorded. Here, the positions of the interference maxima [12] are given by: $y = n\lambda x/d$, where x is the distance between the grating and the detection region, d is the grating period, n is the diffraction order and λ the de Broglie wavelength.

To obtain a high contrast of the interference pattern, it is required to have a narrow wavelength distribution. This is challenging for matter-waves because the wavelength depends on the particles' mass and velocity. The mass can be managed either using chemically pure molecules to begin with, or by pre-selecting the particles in flight, using for example a quadrupole mass-spectrometer [13]. In the latter case it is necessary to start with a negatively charged beam, and neutralize the particles using for example electron photo detachment. Alternatively, a mass resolving detection scheme can be used to post-select the particles - for example in a time-of-flight mass spectrometer [14]. Managing the particle velocities is much more difficult, as they often follow a Maxwell-Boltzmann distribution [15]. The velocity can be selected for example using a mechanical filter [16, 17], however, at the cost of signal intensity. Alternatively, a pulsed source can be used and only the part of the particle bunch is sampled which has the right velocity. Keeping in mind de Broglie's formula it is apparent that the peak separation goes down as the particle mass increases. This sets a limit to the maximal particle mass that can be used with a setup of practicable size.

A near-field configuration, such as a Talbot-Lau interferometer (TLI), allows for a much shorter setup and has several advantages for experiments with heavier particles [18].

3.1. Talbot-Lau Interferometer (TLI)

A near-field interferometer can be built on the Talbot-effect [19] which was discovered in 1836 by Henry Fox Talbot. He observed that when a periodic structure, like a grating, is illuminated with monochromatic light self-images of this structure emerge in multiples of the Talbot distance

$$L_T = \frac{d^2}{\lambda}$$

where d is the grating period and λ the wavelength of the incoming wave. For odd multiples the pattern is shifted by half the period, for even multiples the self-image is exact. There is also a fractional Talbot effect at certain rational multiples of L_T where self-images emerge with a higher spatial frequency than in the original diffraction mask.

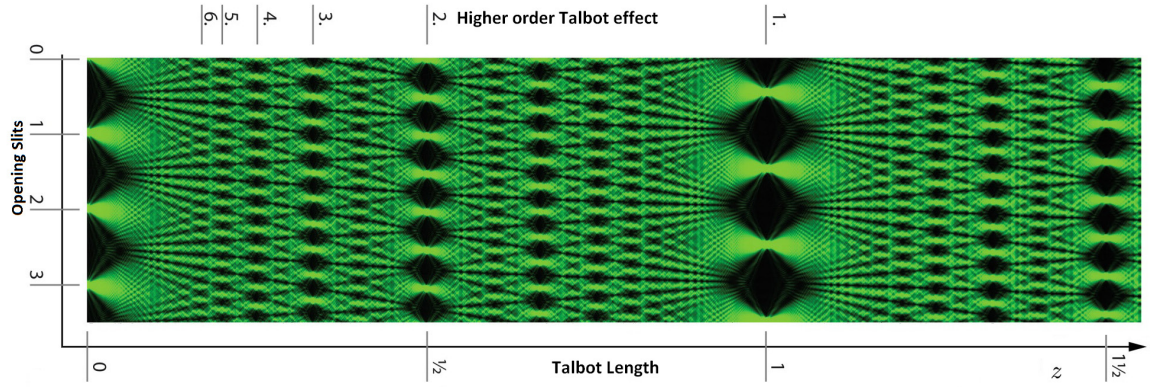


Figure 3.1.1: Talbot effect experimentally measured with light, figure adapted from [20]. A self-image with the same period occurs at the Talbot length. Higher spatial frequency images at rational multiples thereof.

In this setting a spatially coherent beam is still necessary. This can be overcome using the Lau-Effect [21]. In optics the van Cittert-Zernike theorem [22] states that the far-field diffraction image of a coherently illuminated aperture has the same functional form as the coherence function behind that aperture when it is illuminated incoherently. Hence, by adding a second grating in the Talbot distance L_T in front of the diffraction grating the setup can be extended to a Talbot-Lau configuration [23-25]. The first grating acts as a linear array of collimation slits and prepares the coherence for the second grating. Thus, this setup can be operated with an incoherent beam, and collimation slits are in principle no longer necessary. A moderate collimation is, however, still needed to reduce the alignment requirements.

A molecular interference pattern may be observed directly by recording it on a screen (for example a clean Si 111 surface) which is subsequently imaged using a scanning tunneling microscope [26]. Alternatively a third grating can be added in the distance L_T behind the second grating which acts as a scanning mask together with an integrating (non-spatially resolving) detector. Recording the total particle flux as a function of the third grating position allows to resolve the interferometric self-image of the second grating [27].

A setup which can be operated with an incoherent beam utilizes much more of the source signal, since it does not require that much collimation. That gives a significant experimental advantage over far-field diffraction schemes. The main improvement, however, is the scaling behavior. For a given length of the interferometer setup in the near-field the required grating period d scales with the root of λ , while in the far-field d scales linearly with λ . Thus, with increasing particle mass the grating period becomes unpractically small for a far-field scheme much sooner than for a near-field configuration.

3.1.1. Limitations of material gratings

With current nanofabrication techniques it is possible to create millimeter-sized gratings with slit periods down to 100 nm and a periodicity precision of a few angstroms [28].

However, material gratings have a significant limitation: they attract the particles through van der Waals and Casimir–Polder interactions. They introduce a phase shift which has a strong dependence on the particle velocity and the distance of the particle from the wall [25, 29]. These forces are so strong that they can even remove the particles from the beam if they fly close by a grating wall. The interaction potential in the van der Waals regime, near to an extended wall, is given by:

$$U(r) = -\frac{C_3}{r^3}$$

While the interaction potential in the Casimir Polder regime, far from the walls ($\gg$ few 10 nm), is given by:

$$U(r) = -\frac{C_4}{r^4}$$

The interaction constants C_3 and C_4 depend on the gratings' dielectric function and the frequency-dependent polarizability of the particles [30-33]. These dispersive forces pose a problem for highly polarizable and/or slow particles [27, 31, 34]. The deflection induced by the van der Waals forces increases quadratically with the flight time through the grating [35, 36]. It reduces the effective opening fraction of the gratings and, due to the velocity and position dependence, it smears out the interference fringes. A strong velocity selection becomes mandatory, which reduces the useful signal.

There are also more mundane issues, like clogging of the gratings with particles, as cleaning of the gratings is not simple due to their fragile nature.

A solution to these problems is to avoid material gratings. Diffraction structures can be made instead from standing light waves [37, 38]. With narrow-band lasers the grating period can be defined with high precision and by varying the laser intensity it is possible to control the grating transmission function. The particles can interact with such gratings through their optical polarizability coupling to the periodically modulated electrical field and via photon absorption.

One experimental realization of a Talbot-Lau interferometer using a standing laser light wave is the *Kapitza–Dirac–Talbot–Lau Interferometer* (KDTLI) [39, 40]. It uses a phase grating [41] in place of the central diffraction grating. The interaction between the

particles' polarizability and the light field imprints a periodic phase modulation onto the particle beam. This type of grating neither acts as an absorptive mask, nor does it remove particles from the beam. Thus, more signal is transmitted through the apparatus. The heaviest molecule to interfere in this type of interferometer and a long-time record holder is a functionalized 10,123 amu Tetraphenylporphyrin derivative with fluoroalkyl chains attached [42].

As in this type of interferometer the first (coherence preparation) and the third (scanning) grating must be absorptive. Thus, material SiN_x gratings are still used. The van der Waals interactions then continue to pose problems when they become so strong that no particles can pass through the gratings.

One method of implementing absorptive light gratings is to choose a laser with sufficient photon energy to efficiently ionize the particles which pass through the antinodes of the standing light wave [43].

3.2. Optical Time-domain Interferometer for Matter-waves (OTIMA)

To push the mass limits of matter-wave interferometry further, a Talbot-Lau setup with three optical gratings was designed [43, 44]. Such a setup does not suffer from dispersive van der Waals interactions, and the gratings are practically indestructible. By retro-reflecting vacuum ultraviolet (VUV) laser light on a plain mirror, uniform gratings with a period of 78.8 nm can be implemented. Moreover, the grating's opening fraction can be controlled through the laser intensity. With spatially incoherent molecular sources, such an arrangement cannot be realized using phase gratings alone. The first grating must be absorptive in order to prepare the coherence by spatially constricting the particle distribution and enforcing a broad momentum distribution, due to Heisenberg's uncertainty principle. The third grating needs to be absorptive as well, in order to act as a mask to probe the resulting interference pattern. Implementing such gratings for atoms is easy. They can be pumped into dark states, in which they are no longer registered by the employed detection scheme [45, 46]. Larger particles like molecules or clusters of atoms/molecules, however, no longer have the needed state addressability. In the OTIMA setup the absorptive gratings are implemented using ionization induced by single photon absorption in the antinodes of the standing light waves which make up the gratings. VUV lasers with a wave length of 157.6 nm, having a photon energy of ~ 7.9 eV, easily exceed the ionization thresholds of many particles, such as metal/semiconductor clusters, a

number of biomolecules, and van der Waals clusters of molecules. Once ionized, the particles are removed from the beam with a deflection electrode, operated at about 200V. It is placed in a distance of $\sim 20\text{mm}$ from the particle beam so that the neutral particles are not measurably affected.

As an alternative to single-photon ionization also single-photon fragmentation can be used with a wide range of loosely bound particles. One specific class of candidates are weakly bound van der Waals clusters. When they get heated by the absorption of a single VUV photon they may evaporate one or more molecules. The resulting recoil can be sufficient to remove the particles from the acceptance angle of the detector. Another class of candidates can be tailor-made molecules with a predetermined breaking point triggered by the absorption of a photon. Using a mass resolving detector, particles which passed through the grating anti-nodes can easily be distinguished from those which remained intact. Eventually, also doped helium nanodroplets could be used. They can be generated in a mass range between 10^4 and 10^9 amu [47, 48], and due to their low heat capacity (7.2 K/atom) even long-wavelength photons absorbed by a dopant can evaporate many helium atoms.

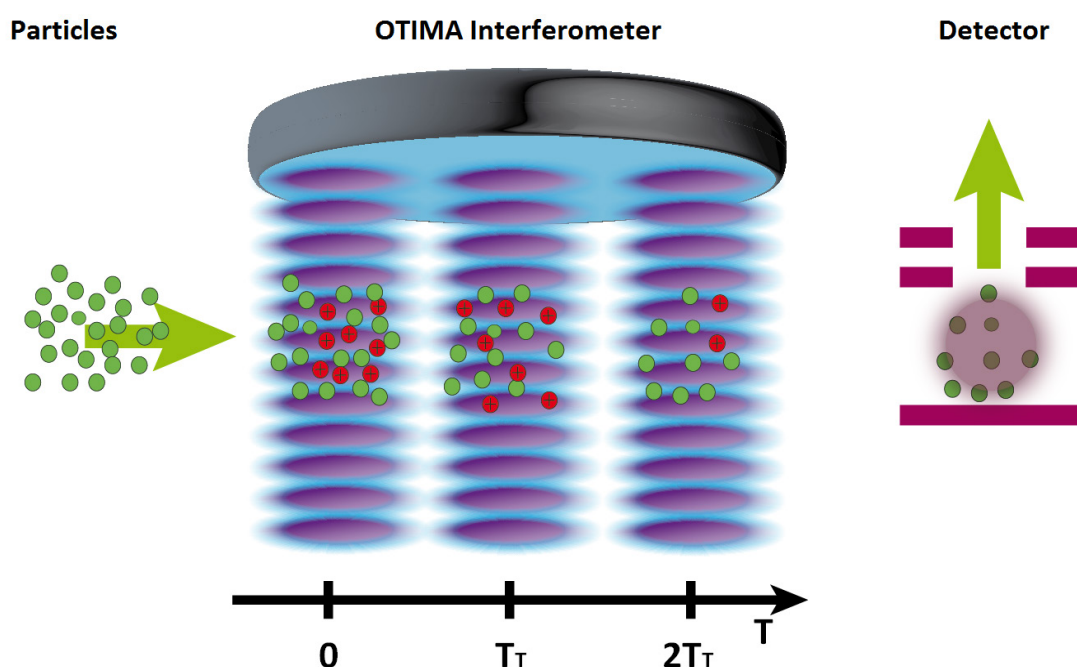


Figure 3.2.1: Schematic depiction of the OTIMA experimental scheme. A pulse of particles enters the interferometer. Some of them get ionized in the antinodes of the first optical grating which thereby prepares the spatial coherence. The matter-waves are then diffracted at the second optical grating and the emerging interference pattern is probed by the third grating. The particles that make it through the interferometer are ionized and detected.

3.2.1. Talbot-Lau interference in the time domain

The OTIMA setup operates in the time-domain [49-53]. Instead of permanent gratings at a fixed position in space, pulsed gratings – sharply defined in time – illuminate a large area. The VUV laser beams have a pulse length of ~ 7 ns and cover an area of about 1×10 mm on the mirror, with the long axis oriented along the particle beam. Pulsed gratings have two advantages: first, it is much easier to precisely control timing than position and second, the interference pattern does no longer depend on the particle velocity. In a regular Talbot-Lau setup the gratings have to be separated by the distance $L_T = d^2/\lambda$ in order for the interference pattern to emerge, where $\lambda = h/(m v)$. Given a particular particle velocity v the self-image of the diffraction grating will appear after a fixed flight time L_T/v once the particles cover the Talbot length. This flight time now defines the Talbot condition and is called the Talbot time:

$$T_T = \frac{L_T}{v} = \frac{m d^2}{h}$$

This criterion no longer depends on the particle velocity and, instead of defining the grating positions in space, it defines the moment in time when the particles of a mass m must interact with the grating in order for the interference pattern to appear. This can be pictured like this: each velocity class of particles will experience its own Talbot-Lau interferometer with the required grating separation in space. Hence, all particles of a given mass show the same interference pattern in the time domain, independent of their velocities. The interference contrast is dependent on the proper Talbot-time T_T , as shown in **figure 3.2.1.1**.

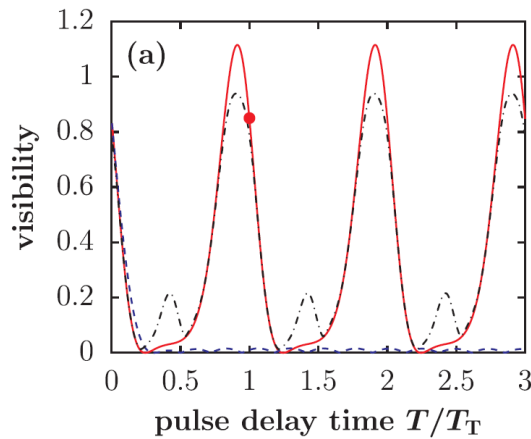


Figure 3.2.1.1: Theoretically computed visibilities as a function of the grating timing. The red line represents the sinusoidal visibility (which is not perfectly accurate), the black dash dotted line the quantum contrast (calculated accurately) and the dashed blue line the classical expected fringe contrast. The figure is taken from [44].

For this interferometer arrangement to work, it is necessary that all particles which reach the detector will have interacted exactly once with each of the three light gratings at the right times. Thus, it is required that the particle beam is also pulsed and while there is no velocity dependence, in principle, there are still some limitations: The pulse length and its velocity distribution must be such that consecutive pulses do not overlap on their way to the detector. Also the grating laser pulses must cover a sufficient portion of the packet along the particle beam such that, even though the packet broadens due to different particle velocities in the pulse, a section of the pulse will consist only of particles which interacted with all of the gratings. This is simulated in **figure 3.2.1.2**, for a particle pulse from a chopped thermal beam. Hence, a detection scheme is required which allows to select these part of the particle pulse. Detecting particles which haven't seen all three gratings will add a substantial background and diminish the interference contrast.

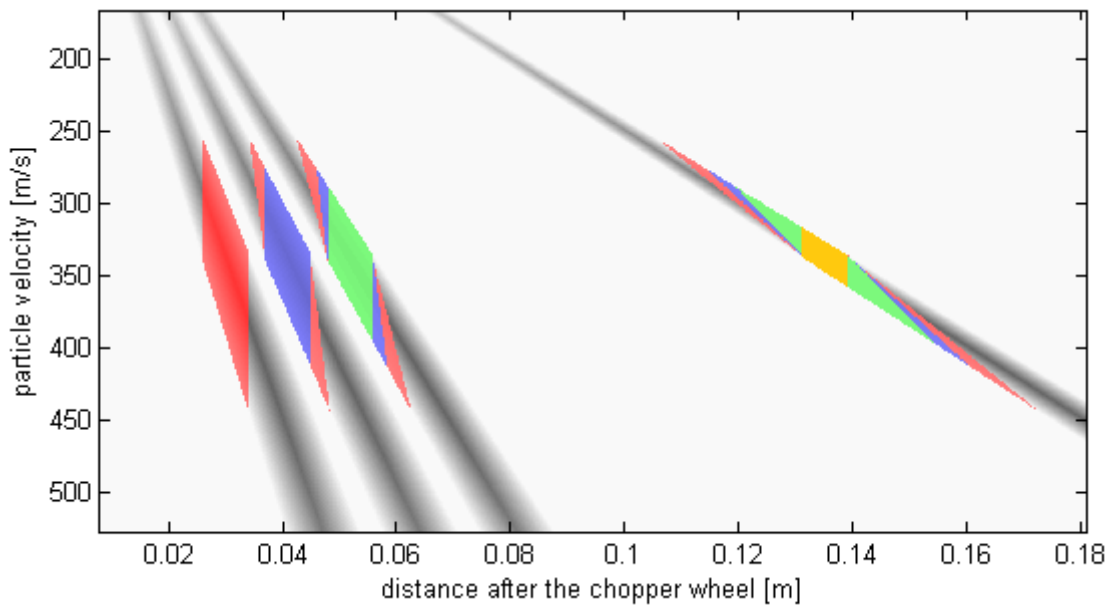


Figure 3.2.1.2: Simulated propagation of a particle pulse emerging from a thermal source after a chopper wheel showing the portion of the beam which interacted with the respective laser gratings and the clean portion of the beam to reach the detector. The laser width here is 8mm and the separation is 11 mm, the detector is placed 80 mm after the interferometer. Each of the gray distributions shows the particle pulse at different moments in time. The red color marks the portion of the beam that interacted with the first laser. The blue region marks the particles that have interacted also with the second laser. Analogously, the green region marks the particles which also interacted with the third laser. Finally, the yellow color shows the part of the pulse which interacted with all three grating lasers once the pulse arrives at the detector.

3.2.2.OTIMA's optical gratings

A full theoretical description of the ionizing time-domain Talbot-Lau interferometer was provided by Stefan Nimmrichter in his thesis [54] and presented in several publications [44, 55]. Here, only a rough overview of his work will be presented.

The optical standing wave interferometer gratings have an intensity distribution given by:

$$I(x, y, z) = \frac{1}{2} c \epsilon_0 E(x, y, z)^2 = \frac{8P}{\pi \omega_y \omega_z} \cos^2 \left(\frac{\pi x}{d} \right) e^{-2 \frac{y^2}{\omega_y^2} - 2 \frac{z^2}{\omega_z^2}}$$

Here the beam is retro-reflected along the x-axis and the z-axis is placed along the particle beam path. The beam profile is assumed to be a Gaussian in y and z and the respective beam waists are ω_y and ω_z . The laser power is P and its wavelength is twice the grating period d .

The particles passing through the interferometer interact with the gratings by photon absorption and through their polarizability which couples to the periodically modulated electrical field. The effects of Rayleigh and Mie scattering are only relevant for particle sizes comparable with the photon wavelength and thus can be neglected [44].

The mean number of absorbed photons $n(x)$ in the grating is dependent on the x position, it is the integral of the absorption rate $\Gamma(x, y, z) = I(x, y, z) \sigma / h\nu$ along the z -axis during the laser pulse length t . Assuming that the particle beam is sufficiently narrow and centered in the maximum of the laser beam the y -axis can be neglected. So $n(x)$ it is given by:

$$n(x) = \frac{\sigma 8Pt}{\pi \omega_y \omega_z h\nu} \cos^2 \left(\frac{\pi x}{d} \right) \equiv n_0 \cos^2 \left(\frac{\pi x}{d} \right)$$

where n_0 describes the average number of photons absorbed by a particle with an absorption cross section σ in the antinodes of the standing light wave.

The photon absorption can be described using Poissonian statistics, the probability that a selected particle absorbs k photons, when a particle on average absorbs q , is given by:

$$P_q(k) = \frac{q^k}{k!} e^{-q}$$

Thus, the probability that a particle absorbs at least one photon for a particular n_0 is given by:

$$p = 1 - P_q(0) = 1 - e^{-n_0}$$

In our setup we usually try to have $n_0 = 8$ giving us a probability of 99.96% that a particle in the grating's antinodes will absorb at least one photon.

Assuming a single photon is sufficient to remove a particle from the beam, the transmission function of one grating can be calculated:

$$t(x) = e^{-n_0 \sin(\frac{\pi x}{d})}$$

With it the transmission through such a grating can be simulated, as done in **figure 3.2.2.1**.

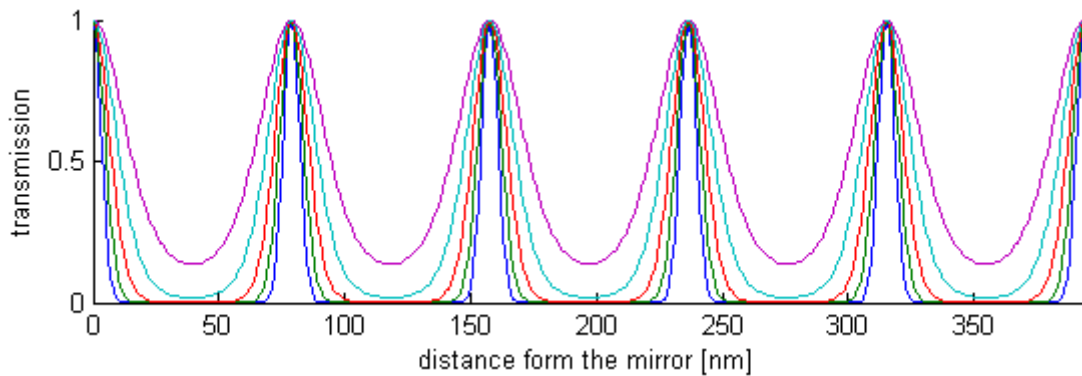


Figure 3.2.2.1: Calculated transmission through one OTIMA grating for different values of n_0 . Magenta: 2; cyan: 4; red: 8; green: 16; and blue: 32 photons.

The interference contrast depends strongly on the quality of the grating. This means that particles entering the antinodes will be reliably removed from the beam, see **figure 3.2.2.2**.

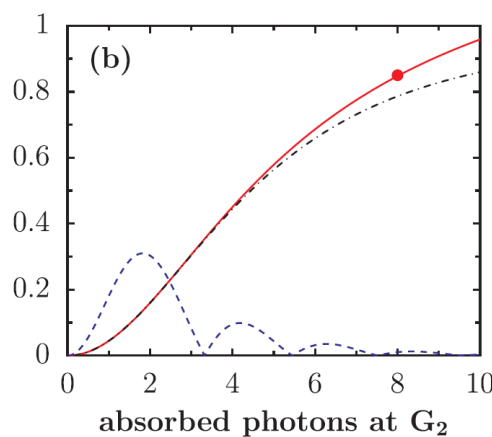


Figure 3.2.2.2: Theoretically computed visibilities as a function of the number of absorbed photons in the second grating at the ideal Talbot time $T=T_T$. The number of absorbed photons in the first and third grating is 8. The solid red line represents the sinusoidal visibility, the black dash dotted line the conventional quantum contrast and the dashed blue line the classical expected fringe contrast. The figure is taken from [44].

The first and last grating in the OTIMA scheme can be realized inversely i.e. the source would provide negatively charged particles which would be neutralized by the first grating. Similarly the ions created in the last grating would not be discarded but supplied to the detector instead of having to ionize the transmitted neutrals. Such an arrangement would however result in a reduced visibility, as shown in **figure 3.2.2.3**.

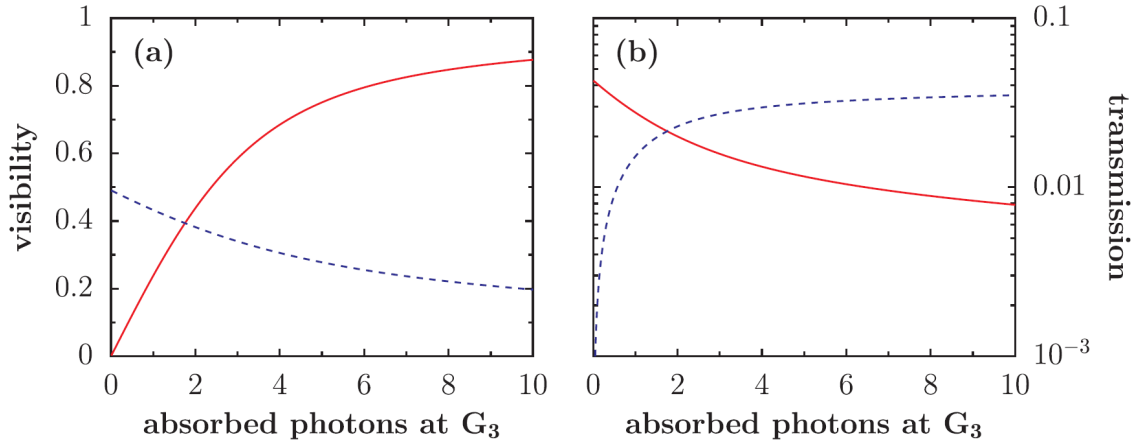


Figure 3.2.2.3: Theoretically computed a) interference contrast and b) transmission through all three gratings as a function of the number of photons absorbed in the third grating. The number of absorb photons in the first and second grating is 8. The solid red line represents the normal case where the ionized particles are removed and the neutrals reach the detector. The blue dashed line represents the inverse case where the ionized particles are detected and the neutrals discarded. The figure is taken from [44].

When polarizable particles cross the standing light wave they experience also a position dependent phase shift [41]. For the first and third grating this is inconsequential. In the middle diffraction grating, however, it influences the interference pattern [54], see **figure 3.2.2.4**.

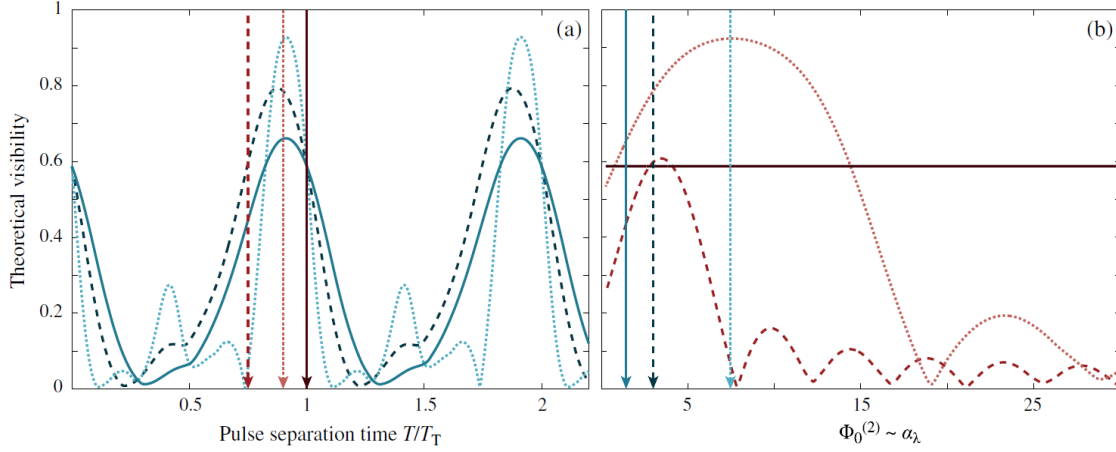


Figure 3.2.2.4: Theoretically computed visibilities using the model [56] for different optical polarizabilities α , at a fixed $n_0=6$. The left figure (a) shows the visibility as a function of the laser grating timing. It is plotted for three different polarizability induced phase shifts in the second grating (dotted line: $\Phi_0^{(2)} = 7.5$, dashed line $\Phi_0^{(2)} = 3$, solid line $\Phi_0^{(2)} = 1.5$). The right figure (b) shows the visibility as a function of $\Phi_0^{(2)}$ which depends on the optical polarizability α . The lines are plotted for different grating timings (dashed line: $T=0.75T_T$, dotted line: $T=0.9T_T$, solid line: $T=T_T$). When the grating timing is perfectly accurate i.e. $T=T_T$ the interference pattern is independent from the particles optical polarizability. The arrows and colors in the figures cross link them to each other. The figure is taken from our publication [56].

The matter-wave acquires a position dependent phase shift while flying through the grating, which can be described with [40]:

$$\phi(x) = \frac{16Pat}{\hbar c \omega_y \omega_z} \cos^2\left(\frac{\pi x}{d}\right) \equiv V_0 \cos^2\left(\frac{\pi x}{d}\right)$$

Here α is the wavelength dependent optical polarizability of the particles. With this, an overall transmission function of the grating can be written down:

$$t(x) = e^{\left(-\frac{n_0}{2} + iV_0\right) \cos^2\left(\frac{\pi x}{d}\right)}$$

The full theoretical descriptions of the matter-wave propagation through the OTIMA interferometer are also given in the theses [57, 58].

4. Experimental setup

The OTIMA interferometer is made up of four sections: a particle source, the interferometer assembly with three laser light gratings, a detector, and a laser beam line, depicted in **figure 4.1**. In order to guarantee a reliable working of the interferometer a number of demanding experimental requirements had to be fulfilled. This are, for instance, the pulse energy and the wavelength of the lasers, the collimation of the beam and the velocity of the molecules.

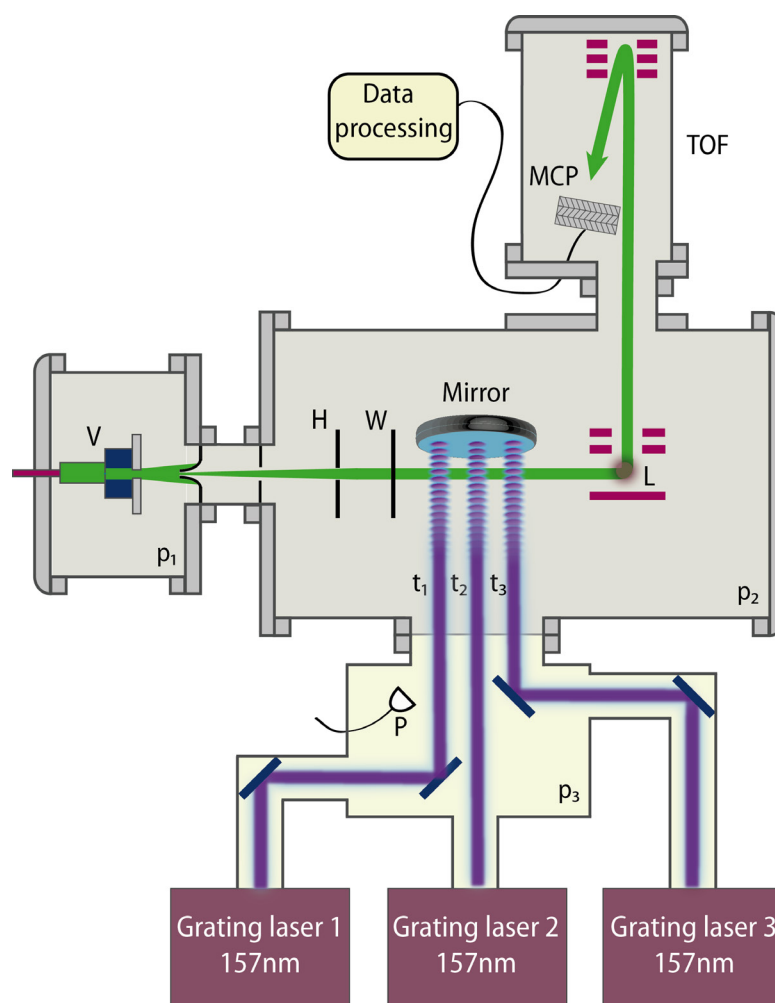


Figure 4.1: Simplified scheme of the OTIMA setup. The Even-Lavie source (left-hand side) was used for interferometry with organic clusters [59]. The particles start in the source chamber ($p_1 \approx 10^{-6}$ mbar) and cross a differential pumping stage. Once the beam enters the main chamber ($p_2 \leq 10^{-8}$ mbar) it is delimited in height and width by mechanical slits and flies across the interferometer to be then ionized and analyzed in the Time-of-Flight Mass-Spectrometer (TOF-MS). The three VUV laser beams, which make up the matter-wave interferometer, are enclosed in an evacuated and N_2 purged beam path ($p_3 \approx 1$ mbar) to ensure a high transmission from the laser to the vacuum chamber. The figure is taken from our publication [59].

The pulsed nature of the OTIMA interferometer suggests that the particle beam should be pulsed as well to ensure that all particles have seen the same laser pulse sequence.

This is achieved in a source with a pulsed opening or a subsequent chopping mechanism. The length of the interferometer constrains the maximal particle velocity for a given Talbot time. Hence, massive particles should emerge already slowly from the source or they must be further cooled later, as described in **chapter 4.1**.

The photon energy in all three OTIMA lasers must be sufficiently high to deplete the particle beam by single-photon absorption. And the number of photons (laser intensity) must be sufficient to deplete the particle beam in the antinodes of the standing light waves. The laser timing must be accurate enough to fulfill the conditions for cluster interference in the time domain, see **chapter 3.2** for details. The short laser wavelength of 157 nm and energies of ~ 5 mJ/pulse can currently only be obtained with F₂ excimer lasers and also the pulsed timing requirement (jitter < 5 ns) is fulfilled by our three EX50 GAM lasers. Their beam paths must be oxygen-free on the level of 1 ppm, since oxygen strongly absorbs at this wavelength.

After passing the interferometer the particles are detected by an orthogonal reflectron Time-Of-Flight Mass Spectrometer (TOF-MS)². We run the experiment at a repetition rate of 100 to 250 single shots per second, mostly limited by the maximal repetition rate of the excimer lasers. This high repetition rate necessitates a sophisticated data acquisition and management scheme. The experiment is controlled by the custom made software MOPS (*Molecular Optics Programming System*) that I have developed and optimized in particular for the OTIMA experiment. This software controls all experimental timings by pulse/delay generators. It is also capable of controlling additional hardware, like flow regulators and lasers. It records, processes and stores the mass spectrometer signal acquired by a high speed digitizer card in real time.

² Bipolar Reflectron TOF model RFT50, Fa. Stefan Kaesdorf München

4.1. Molecule and cluster beam sources

A challenging task in matter-wave interferometry is the development of sources that are sufficiently universal to be used for a variety of different particles. The velocity distribution of these particles must in particular be compatible with the interferometer constraints. The pulsed optical gratings in the OTIMA interferometer have a period of 78.8 nm, thus the required Talbot time for a particle of a mass m is $T_{\text{Talbot}} = 15.4 \text{ ns} \cdot m \left[\frac{1}{\text{amu}} \right]$, see **chapter 3.2** for details. The particles must travel the distance between two gratings in this time, in order to interact with all three light gratings while passing through the interferometer. Because the laser beam has a finite width along the particle beam path, in our current setup this is 10 mm, this imposes a constraint on the maximal velocities that can be handled by the particular apparatus. **Figure 4.1.1** shows the extreme combinations of particle mass and velocity that can be handled by the OTIMA setup.

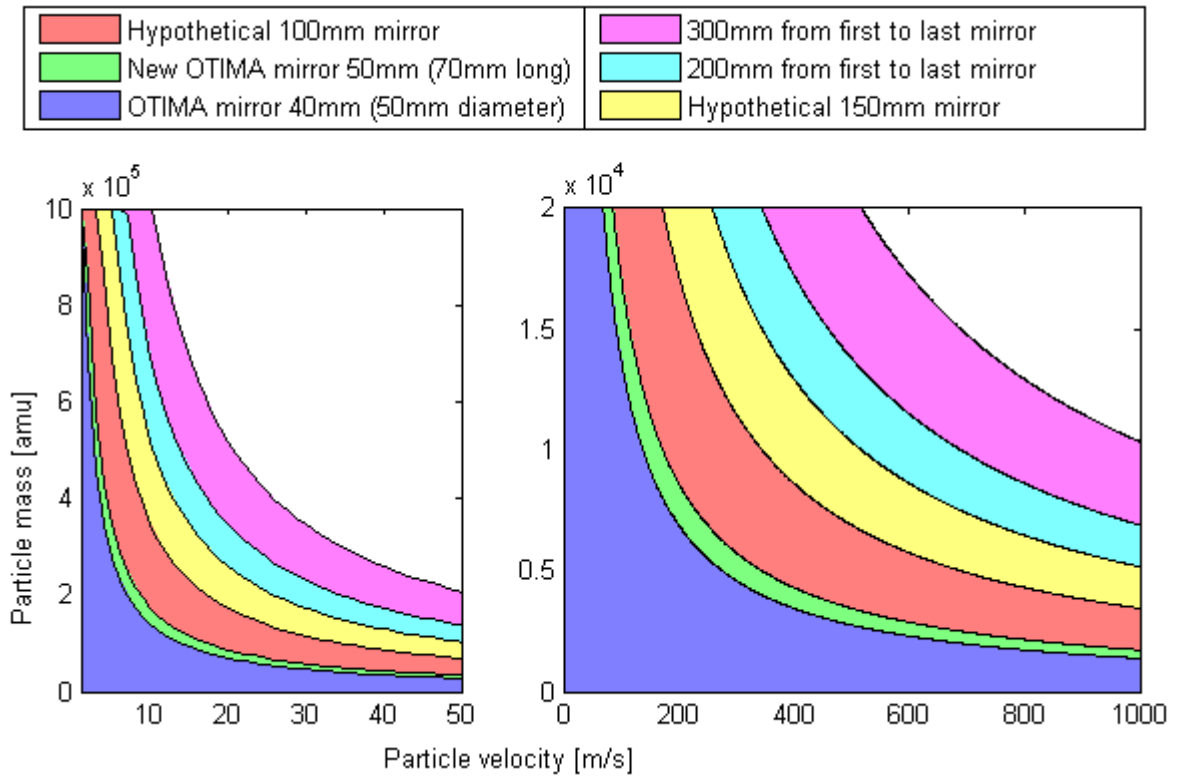


Figure 4.1.1: Maximal particle mass compatible with first order OTIMA interference as a function of the velocity for various grating separations. The currently installed circular 50 mm mirror (blue) will be replaced with a rectangular 70×40 mm mirror (green) with a usable area of 50×20 mm. The main advantage of the mirror will be a better surface quality. Additionally, due to its size it will also allow to handle slightly heavier particles. For example, at 75m/s the old mirror could go up to 18,400 amu while the new one can reach 23,000 amu, and at 50m/s its 27,500 amu for the old one and 34,400 amu for the new one. Since much longer grating separations are possible in a future setup their values have also been plotted.

Not only the velocity but also the length of the particle bunch must be selected in order for particles from adjacent pulses not to overlap. For Talbot times up to $100\text{ }\mu\text{s}$ the packet length should be shorter than $100\text{ }\mu\text{s}$.

The particles should also be *internally* sufficiently cold not to emit thermal photons or cluster fragments along the way. Otherwise they would disclose the path of the particle through the interferometer (see **figure 4.2.1** in **chapter 4.2**) and therefore cause decoherence [60-63]. We also prefer to work with neutral objects, since it is difficult to shield slow ions from electromagnetic perturbations that would easily shift the particle position and smear out the interference pattern [64, 65].

The requirement of sufficient beam flux is non-trivial and in recent years we have explored a number of different source types, which will be described in detail in the following sections.

4.1.1.1. Magnetron sputter source

We first tested a magnetron sputter source [66], which was provided by Prof. Bernd von Issendorff from the Albert-Ludwigs Universität in Freiburg. It can generate intense beams of metal and semiconductor particles, ranging from single atoms to cluster masses beyond 10^6 amu. It consists of a sputter head where atoms and small particles are released from a bulk target to further condense in an aggregation tube to larger clusters.

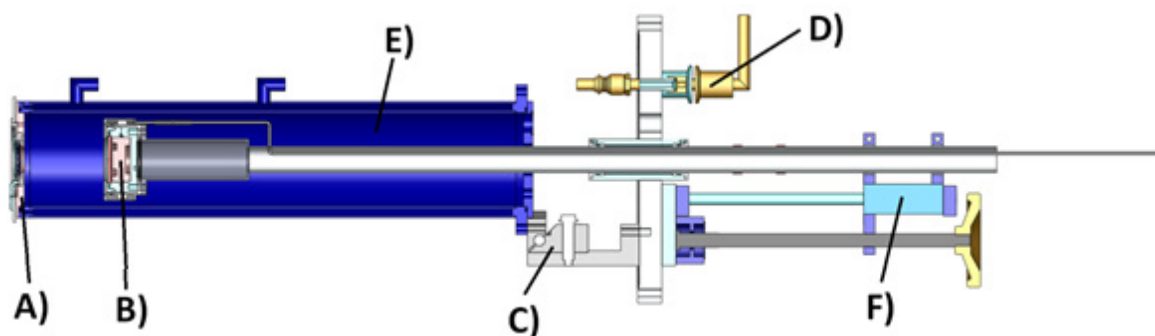


Figure 4.1.1.1: Magnetron sputter source: A) exit aperture with a variable iris, B) sputter head, C) hinge allowing to adjust the source position, D) liquid nitrogen (LN₂) feed through, E) LN₂ cooled condensation tube, F) translation mechanism that allows moving the sputter head.

The source ignites an argon plasma over a metal or doped semiconductor target which has a diameter of 50 mm and a maximum thickness of 5 mm. The target is connected to a DC power supply³ while the nozzle ring (**figure 4.1.1.2 part A**) that supplies the argon gas is connected to ground. The supply is set to keep the power constant around a value typically within 25-100 W. A permanent magnet beneath the target guides the electrons in helical trajectories to propagate along the magnetic field lines. The radii of these trajectories are determined by balancing the Lorentz force, acting on the charged particles, and the centripetal force. These Larmor radii are proportional to the particle mass m and its velocity perpendicular to the magnetic field lines $v_{\perp} = \sqrt{2qU/m}$. Here q is the particles' charge and U the potential applied by the power supply to the target with its base (**figure 4.1.1.2 components B, C, D, F**) while the nozzle ring (**figure 4.1.1.2 component A**) remains on ground potential. From this we get: $r_g = \frac{mv_{\perp}}{|q|B} = \sqrt{\frac{2mU}{|q|B^2}}$. Hence, electrons will have radii of about 100 μm , and singly charged argon ions will circle with a radius ≥ 100 mm. The ions are therefore mostly unaffected by the magnetic field and can be considered to impact the sputtering target in “free fall” [67]. The permanent magnet

³ Advanced Energy MDX 500

which provides the B field is shaped with its south pole in the target center and the north pole around its circumference. The magnetic field is weakest between these poles. This means that in this region the Larmor radii for the electrons are largest, and therefore the electron density highest. Subsequently, the discharge is concentrated in a ring near to the target surface.

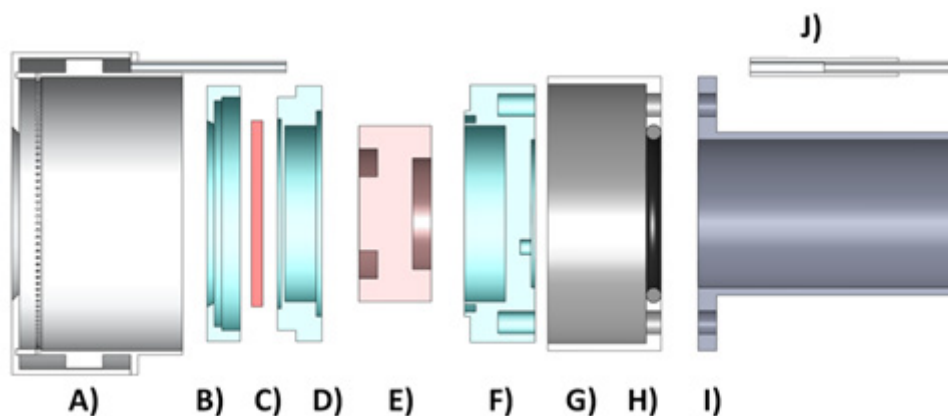


Figure 4.1.1.2: Magnetron Sputter head: A) nozzle ring on ground potential, B) target holder ring, C) sputtering target, D) target back plate on power supply potential, E) magnetron magnet, F) magnet holder, G) PTFE isolator, H) viton sealing, I) mounting tube, J) gas supply.

The ion energy, typically a few hundred eV, exceeds the threshold for sputtering, which is approximately four times the sublimation energy for the bulk material [68, 69]. Therefore, not only single atoms are sputtered off the target, but also dimers, trimers, and larger pieces [70]. The sputtered particles are cooled by collisions with the surrounding buffer gas and aggregate to form larger clusters. The buffer gas is usually a mixture of argon and neon, or argon and helium, cooled to a temperature of 77 K using liquid nitrogen around the aggregation tube. The argon is supplied via the sputtering head at a flow rate of up to 100 standard cubic centimeters per minute (sccm). The neon, or helium, is supplied directly to the aggregation tube at a flow rate between 0 and 500 sccm. The clusters grow [71] inside the aggregation tube at a pressure in the range of 0.1-1 mbar. They then exit through an iris that can be varied in aperture between 1-28 mm, also during operation. The pressure in the vacuum chamber outside the aggregation tube is around 10^{-3} mbar. Under these conditions the clusters do not grow any further. The final cluster size depends on the gas mixture, the pressure inside the aggregation tube, as well as on the distance between the sputter head and the exit aperture, as it changes the cluster aggregation time. We have successfully produced broad cluster distributions using various metal (**figures 4.1.1.3 – 4.1.1.5**) and also semiconductor (**figure 4.1.1.6**) targets.

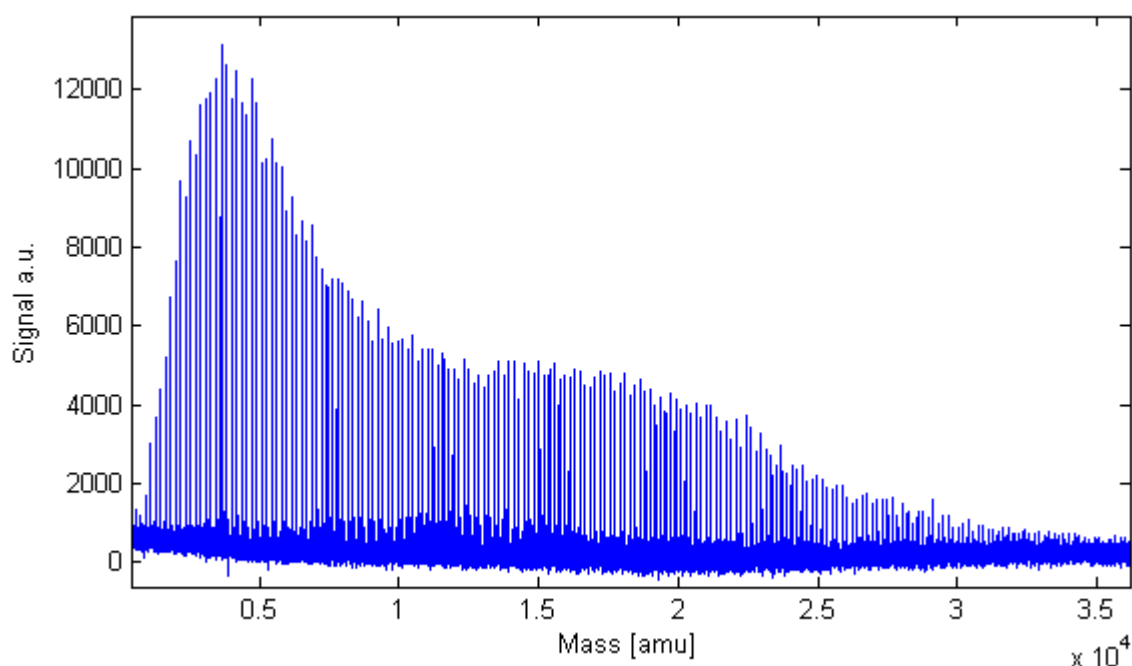


Figure 4.1.1.3: Mass distribution of tantalum clusters ($m_{\text{Atom}}=180.9$ amu) emerging behind the magnetron sputter source. We see single charged clusters composed of up to 175 atoms and some doubly charged clusters as well.

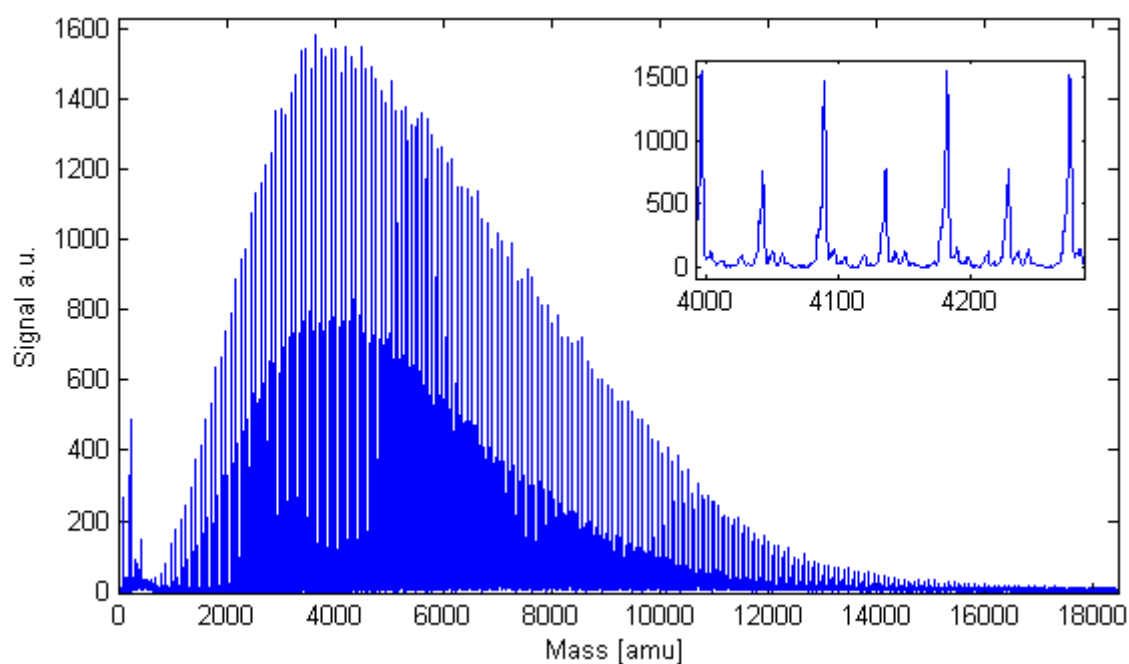


Figure 4.1.1.4: Niobium ($m_{\text{Atom}}=93$ amu) mass distribution with singly positively charged clusters up to $n=170$. The strong signal related to doubly charged clusters is interlaced with the “single”-spectrum, the inlay shows a magnification of the 4000-4300 amu range with four “single”- peaks and three “double”- peaks in between.

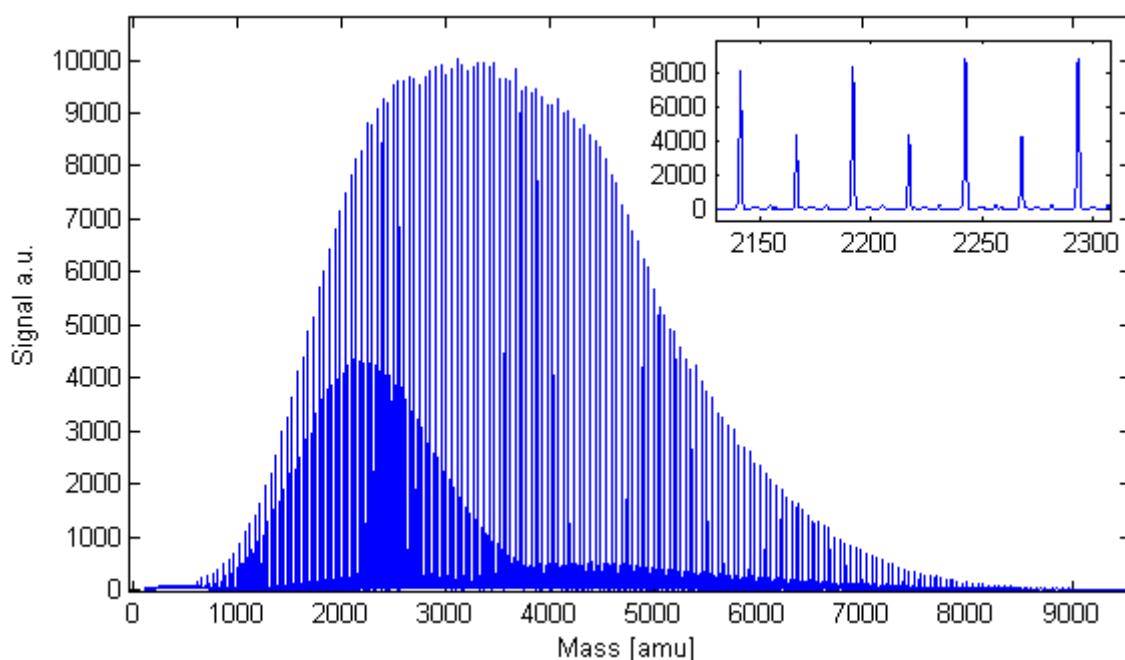


Figure 4.1.1.5: Distribution of vanadium clusters ($m_{\text{Atom}}=50.9$ amu) up to $n=150$. The singly charged clusters are predominant, but we also see a strong signal from double charged clusters, see inlay with magnification. The expected cluster mass distribution should follow a log-normal distribution, and for the doubly charged clusters it does. For the singly charged particles, however, it appears not to do so. This is due the discriminator method used, which has not distinguished between single ion impact events and multiple ion impact events, resulting in losing some of the strongest signal.

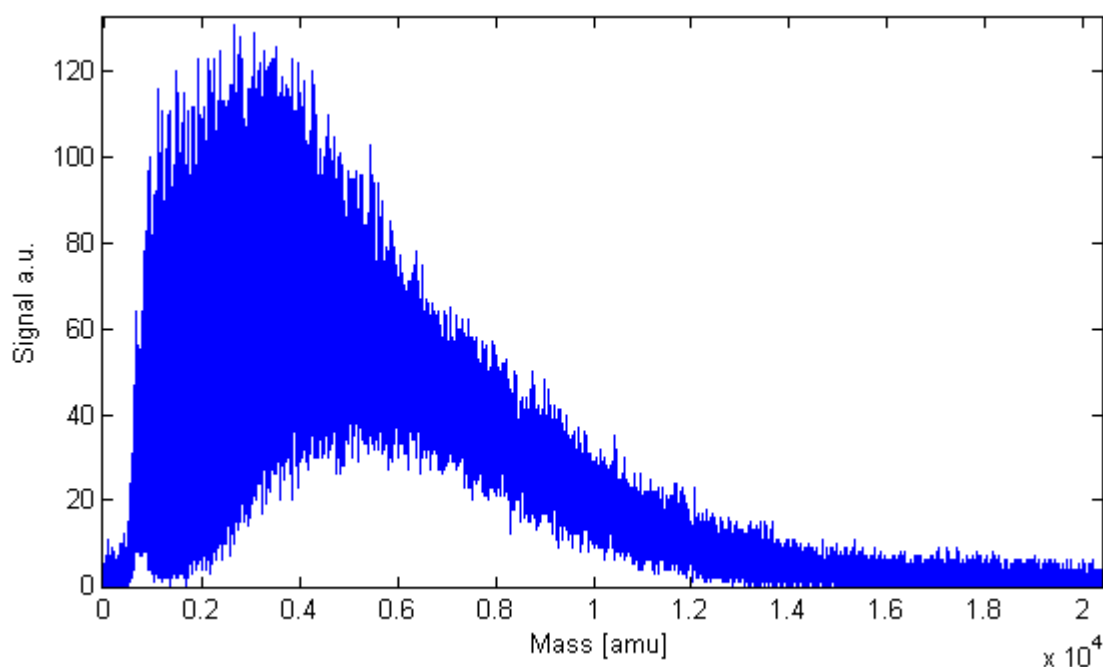


Figure 4.1.1.6: Silicon ($m_{\text{Atom}}=28$ amu) cluster distribution showing clusters up to the $n=500$. The small mass separation does not allow resolving the individual clusters.

The magnetron sputter source produces 1/3 neutral 1/3 positive and 1/3 negatively charged clusters [67]. In our setup we have discarded the charged particles using a deflection electrode which was placed in the first differential pumping stage after the source. The vacuum setup with the two differential pumping stages is presented in **chapter 4.2** and shown in detail in **figure 4.2.2**.

4.1.1.1. Pulse chopping and cluster velocity measurements

The sputtering source provides a constant flux of particles which is not well suited for OTIMA's pulsed mode of operation. Hence, the beam was chopped into packets by a 150 mm diameter wheel rotating at a frequency between 25 and 62 Hz. The wheel had 4 rectangular openings, each 1×3 mm large. Therefore, only about 1 % of the signal could actually be used.

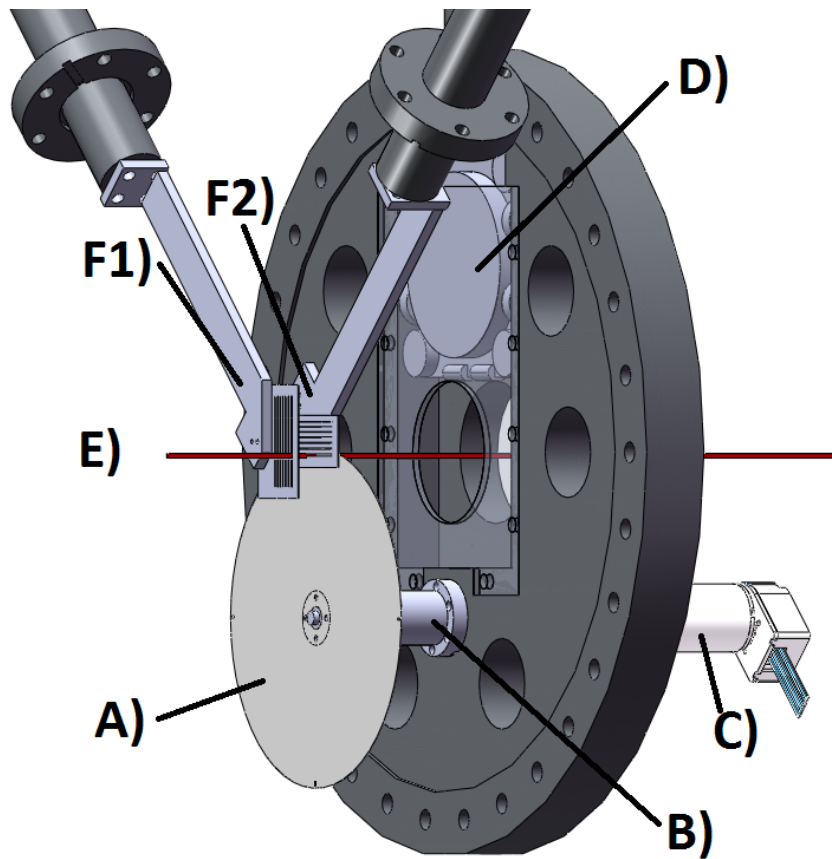


Figure 4.1.1.1.1: Chopper assembly inside the main chamber in front of the interferometer: A) chopper disk with four small slits around the outer circumference, B) ferromagnetic fluid sealed rotation feed through, C) motor, D) built-in valve to separate the main chamber from the source part, E) particle beam, F_{1,2}).vertical and horizontal delimiter slits.

We used the chopper to measure the velocity distribution of the particles by varying the delay between the chopper opening and the detector trigger. We observed a significant velocity slip [72-75] between small and large clusters, i.e. the smaller clusters are faster than the larger ones (see **figures 4.1.1.1.2 – 4.1.1.1.4**). The massive clusters are cooled down in the condensation tube and reach a low thermal velocity. While the carrier gas transports them through the iris they are accelerated by collisions with the fast flowing gas. The collision cross section grows quadratically with the particle radius while their mass grows with the radius cubed. Therefore, assuming rigid sphere collisions, smaller clusters will be accelerated quicker than large ones [76-78].

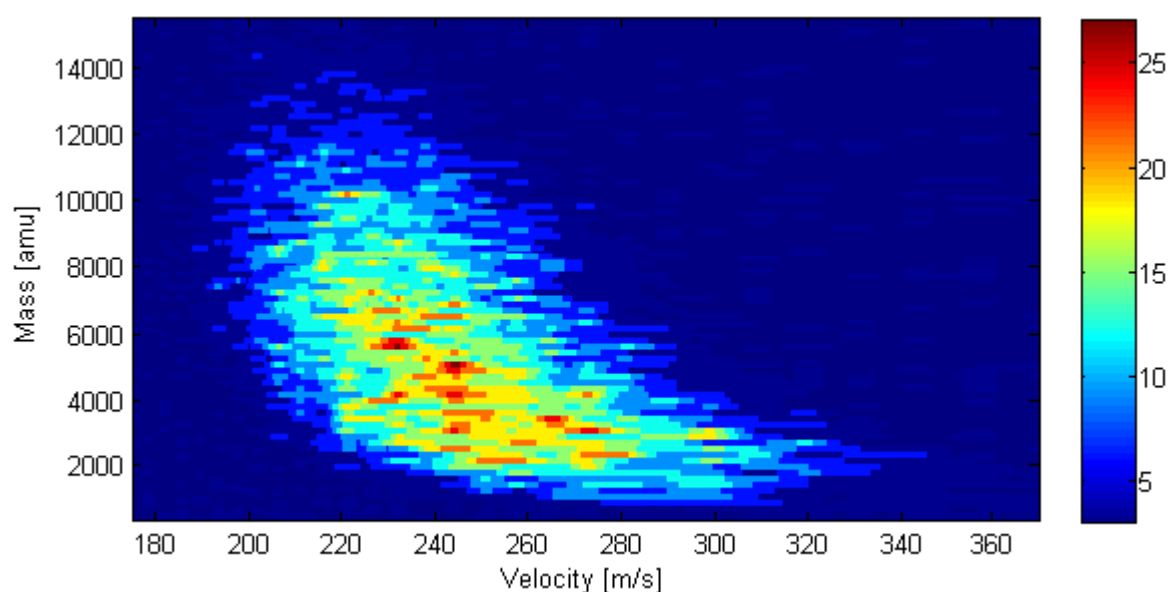


Figure 4.1.1.1.2: Velocity distribution of tantalum clusters emerging from the magnetron source. The color represents the beam intensity in arbitrary units. Source settings: Argon flow at 96 sccm; Neon flow of 344 sccm; Sputter power: 43W

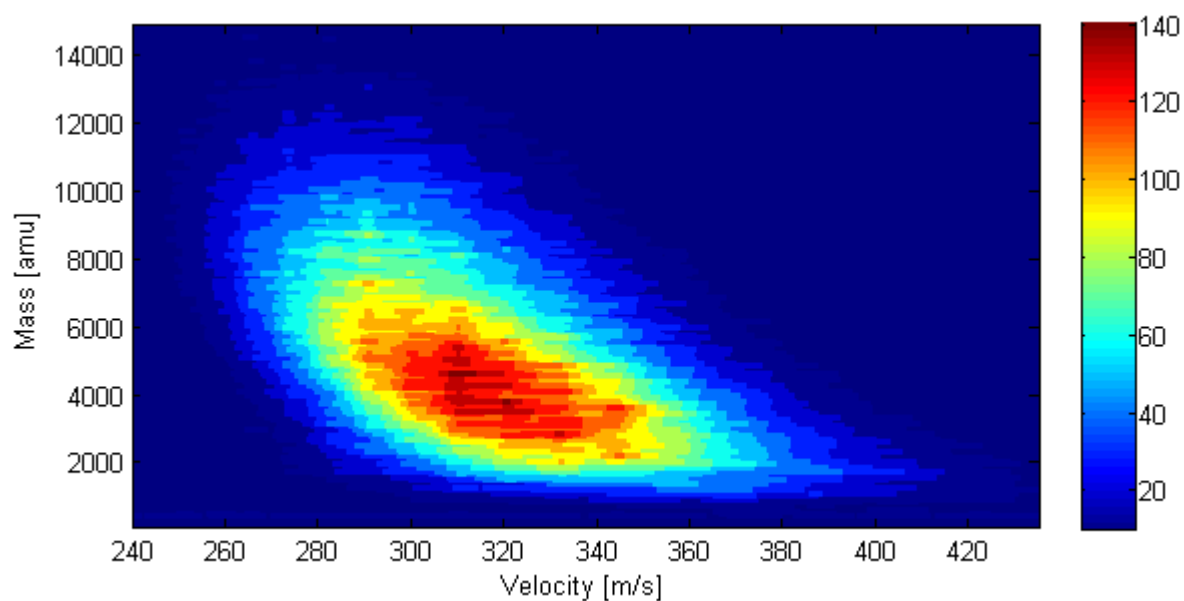


Figure 4.1.1.1.3: Velocity distribution of Nb clusters. The colors represent the beam intensity in arbitrary units. Source configuration: Argon flow: 109 sccm; Helium flow: 510 sccm; Sputter-Power: 35W.

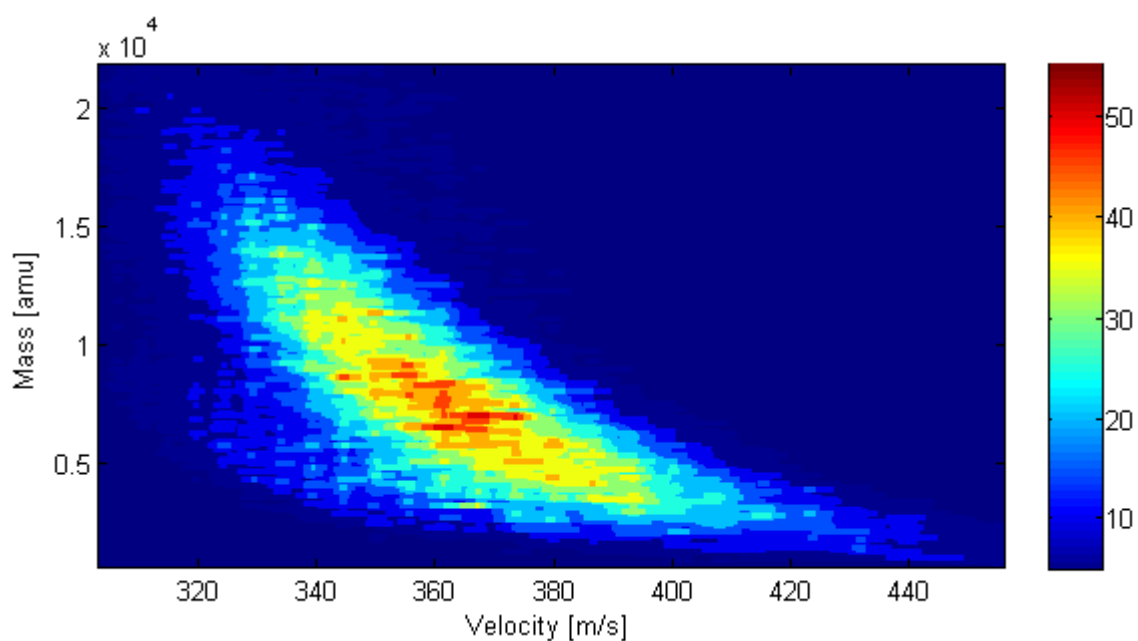


Figure 4.1.1.1.4: Velocity slip in the Vanadium cluster distribution. Source setting: Ar: 41 sccm; He: 348 sccm; Sputter power: 65W.

4.1.1.2. Buffer gas cooling of clusters

In order to slow the clusters further we additionally used a buffer gas cell [79, 80]. It was thermally connected to a pulse tube cooler⁴ with two cooling stages; one at 65 K with 30 W cooling power, the other one at 4.2 K with 0.5 W of cooling power. The pulse tube cooler has smaller vibration levels than a *Gifford McMahon* cooler and in contrast to a liquid helium cryostat it can run continuously with hardly any servicing. The buffer gas cell was equipped with a heating element in order to control the temperature and to prevent the buffer gas from condensing.

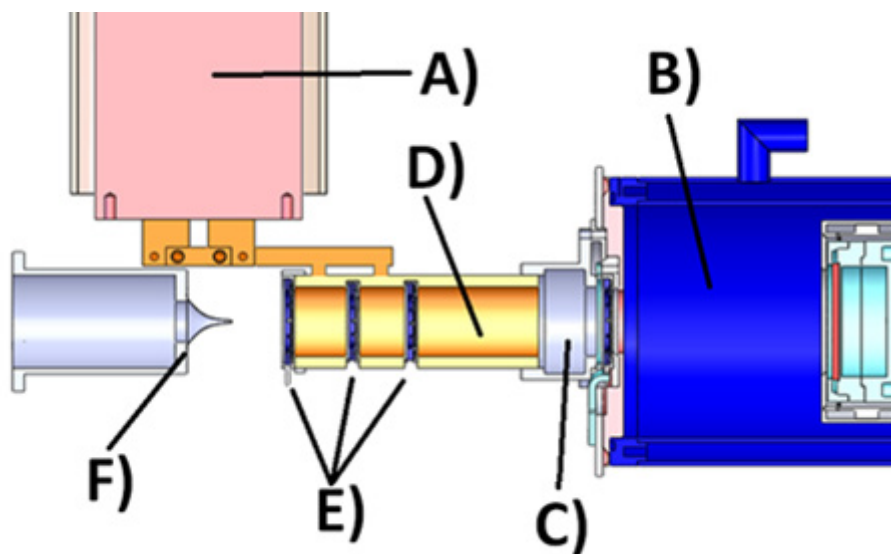


Figure 4.1.1.2.1: Buffer gas cell assembly: A) pulse tube cooler, B) magnetron sputter source, C) PTFE adapter for the buffer gas cell, D) buffer gas cell, E) adjustable irises, F) skimmer. The buffer gas cell is 105 mm long and 30 mm wide, the typical pressure inside is in a range of 0.1-1 mbar.

The clusters reach thermal equilibrium with the buffer gas through collisions. The most probable velocity in an effusive thermal beam with Maxwell-Boltzmann distribution is given by $v_{mp} = \sqrt{2k_b T/m}$. We therefore expect the following gas velocities:

Temperatures:		4K	27K	77K	87K	121K	165K	300K
Gas	Mass	Gas velocity [m/s]						
He	4.00u	130	335	566	601	708	828	1116
Ne	20.18u		149	252	267	315	368	496
Ar	39.95u				190	224	261	353

Table 4.1.1.2.1: Noble gas velocities in m/s at different temperatures, no entry indicates that the gas at that temperature has condensed.

⁴ SHI Cryocooler MODEL SRP-062B

The buffer gas cell is attached after the iris at the end of the condensation tube. It was also equipped with three additional irises, which allow control of the pressure and flow inside the cell [81-83]. This is important as the requirements for cooling are different from those for condensation. The irises also allow better guiding of the gas flow through the cell. They can be operated with a wobble stick while the source is operating, which allows for easy tuning.

When the clusters leave the source they cross three differential pumping stages. The first two are separated by 1 mm skimmers, while the third one has a 3 mm entrance hole. This is necessary as the pressure in the source chamber is typically around 10^{-3} mbar during operation while the main chamber must be kept at 1×10^{-8} mbar or better.

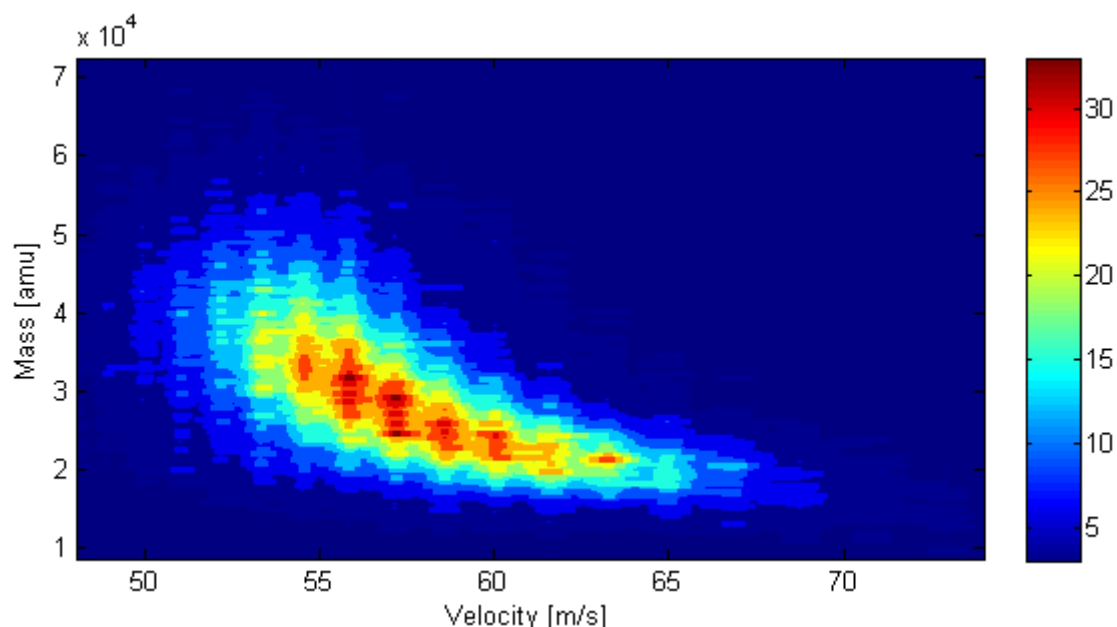


Figure 4.1.1.2.2: Cryogenic buffer gas cooling of Niobium clusters. Source configuration: Ar: 150 sccm; Ne: 35 sccm; Sputter power: 100 W; Temperature of the buffer gas-cell: 24K.

With argon and neon as a carrier gas while cooling the buffer gas cell down to 24 K we reach velocities as slow as 57 m/s for clusters around $m=26,000$ amu, as shown in **figure 4.1.1.2.2**. This is a large improvement in comparison to the normal velocity distribution shown in **figure 4.1.1.1.3**. The produced cluster distribution contains much larger clusters, but the signal intensity is substantially lower. The reached velocities are sufficiently slow for our current interferometer dimensions. Unfortunately, the signal intensity was too low for immediate interference measurements.

4.1.1.3. Native source pulsing

In an attempt to improve the signal strength we tried to pulse the source by placing a fast ball valve - a rotating ball with a hole - at the exit iris. The ball was fixed to the shaft of a DC motor and rotated with 100 Hz, thus resulting in a pulse frequency of 200 Hz. The detection was synchronized to the ball with an optical switch located perpendicular to the particle beam. The ball had a diameter of 40 mm and an opening of 8 mm, hence the duty cycle (ratio of time open to time closed) of the valve was $\sim 1:8$.

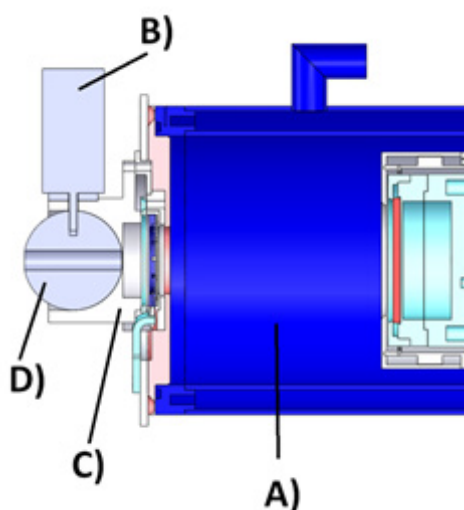


Figure 4.1.1.3.1: Ball valve assembly: A) magnetron sputter source, B) motor, C) ball valve adapter, D) rotating ball with opening.

The average pressure inside the vacuum chamber did not change since the gas flow into the condensation tube was controlled by two mass flow controllers⁵. Thus, while periodically opening/closing of the exit aperture leads to pressure oscillations inside the condensation tube, the amount of gas released into the vacuum chamber remains constant.

⁵ Aera FC-7700CD

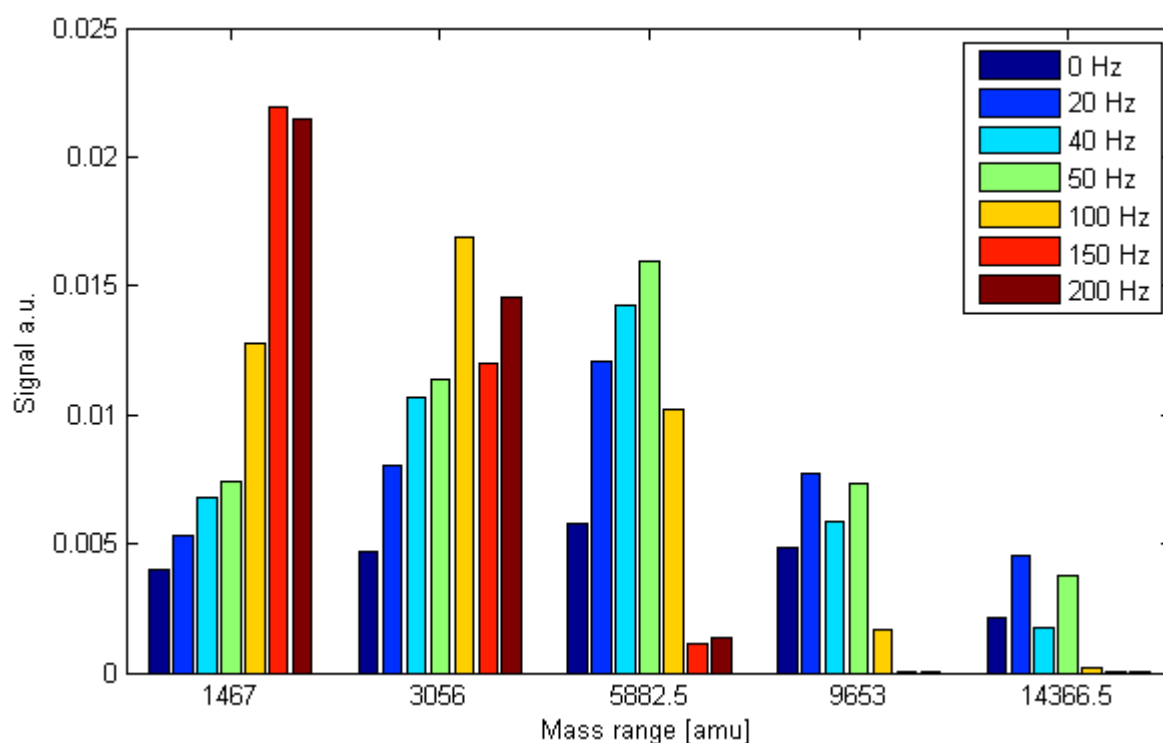


Figure 4.1.1.3.2: Effect of the ball valve on cluster intensity: Cluster signal with the ball valve for 5 selected mass ranges at different speeds including no rotation (permanently open) for reference.

With this modification we achieved a significant signal increase, see **figure 4.1.1.3.2**, for small masses and still some improvement for higher masses. We also observed that at higher pulse rates the signal of heavier clusters was significantly reduced, which is less suitable for our experiments.

4.1.2. Even-Lavie valve

The Even-Lavie valve [84, 85] is a commercial⁶ source that evaporates material into a noble gas and releases both into a supersonic molecular beam through a fast electromagnetic valve. The opening time can be as short as 20 μ s and below, for repetition rates up to 1 kHz. The valve encloses a container which can be heated up to 520 K, where solid or liquid materials can be evaporated. The standard valve did not heat uniformly and made it difficult to reach the target temperature. To compensate for this we added an additional heating element (see **figure 4.1.2.1** part E) to the middle of the vessel, which solved this problem.

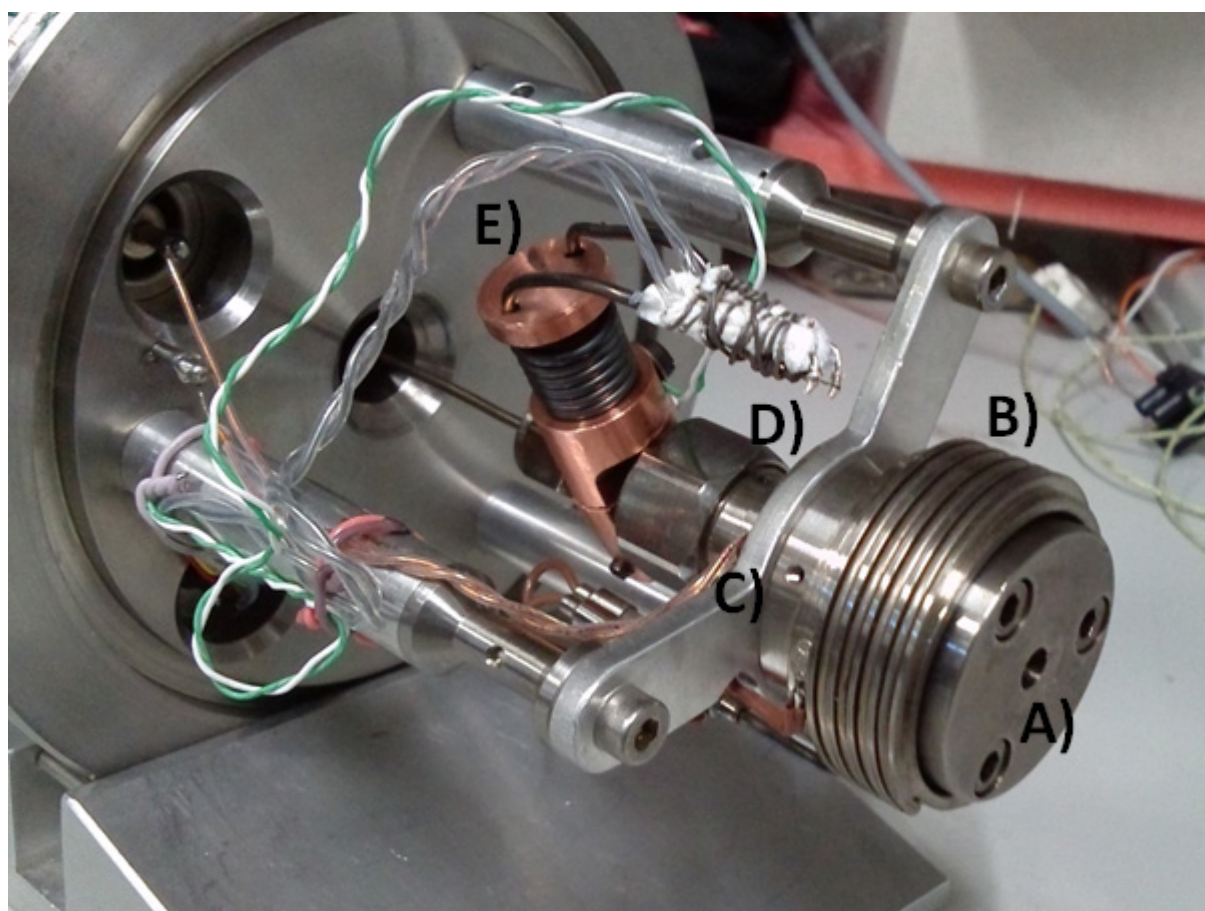


Figure 4.1.2.1: Modified Even-Lavie source: A) nozzle, B) heating wire wound around the valve mechanism, C) valve mechanism, D) particle container, E) custom heating element.

The molecules – in our case anthracene, hexafluorobenzene, caffeine, vanillin etc. – are evaporated in the presence of a seed gas at a pressure between 1-100 bar. We have experimented with most noble gasses, like He, Ne, Ar, Kr, Xe and we also used mixtures

⁶ Nachum A. Lavie, LAMID LTD

of these. The particle-loaded seed gas is periodically released into the vacuum chamber through a 50-150 μm aperture. During this process the particles cool down and form weakly bound van der Waals clusters. The precise composition of these clusters depends strongly on the source settings. The nozzle exit is parabolically shaped to increase the on-axis beam intensity and to enhance cluster growth [86]. In earlier experiments by other groups, Aniline and Anthracene molecules were observed with a rotational temperature of ~ 0.4 K when seeded in He gas [84]. However, the cluster velocity remains high. The terminal velocity of a supersonically expanding gas is given by

$$v_t = v_{mp} f_c = \sqrt{\frac{2K_b T}{m}} f_c.$$

Here, v_{mp} is the most probable thermal velocity and f_c is a dynamical factor [87] which arises through the aerodynamic interaction of the particles during the expansion. It is given by

$$f_c = \sqrt{\frac{\gamma}{\gamma - 1}},$$

with $\gamma = (f + 2)/f = 5/3$ the adiabatic coefficient for monatomic gases with $f = 3$ the number of degrees of freedom per particle. This results in $f_c = \sqrt{2.5} \approx 1.6$. In **table 4.1.2.1** the most probable velocity of various gases in a supersonic beam is shown.

Temperatures:		4.1 K	27.1	87.1	121 K	165 K	300 K	400 K	520 K
Gas	Mass	Gas velocity [m/s]							
He	4.00u	208	537	962	1134	1325	1786	2060	2352
Ne	20.18u		239	428	505	589	795	918	1047
Ar	39.95u			304	359	419	565	652	744
Kr	83.79u				247	289	390	450	513
Xe	131.3u					231	311	360	410

Table 4.1.2.1: Supersonic beam velocities in m/s calculated for different noble gasses at different temperatures, no entry indicates that the gas at that temperature has condensed.

Collisions with the seed gas atoms accelerate the clusters until they asymptotically approach the velocity distribution of that gas. However, for low seed gas pressures the collision rate is reduced and the center of mass velocity distribution does not reach the gas velocity. This is commonly also referred to as velocity slip [88, 89]. When operating our source at 500 K with a seed gas pressure of around 10 bar we observe cluster

velocities of 640 m/s for argon and 960 m/s for neon. Mixtures of these gasses enabled us to prepare velocities in between these values.

At this high velocity our present interferometer setting is compatible with particles up to ~2,600 amu. The high brightness of this source makes it a well suited tool for testing and adjusting the interferometer. With it a single interference measurement can be as short as 10 minutes. The source can operate with a wide range of thermally stable molecules.

4.1.3. Laser desorption

Laser desorption is a versatile tool for bringing fragile particles into the gas phase [13, 90, 91], often with minimal fragmentation. The simplest approach is to directly heat the sample using a continuous laser beam. This excites the molecules thermally by distributing the absorbed photon energy amongst their internal degrees of freedom. The vibrationally excited molecules now have enough energy to overcome the van der Waals or dipole-dipole interactions with the surface and are volatilized. However, direct heating can also cause thermal dissociation, if the particles are not thermally stable, which is often the case for large molecules.

One may prevent this by mixing the molecules of interest with a matrix that is more volatile, more absorptive and more stable. Matrix Assisted Laser Desorption (Ionization), MALD(I) [92-94] brings neutral analyte and matrix molecules as well as their protonated/deprotonated ions into the gas phase .

Another desorption mechanism is based on Laser Induced Acoustic Desorption (LIAD) [95]. This scheme uses a 10-100 μm foil as a substrate for the molecules of interest. A short-pulse desorption laser beam incident on the back side of the foil liberates molecules from the front side. Typical laser pulses may be several nanoseconds long and have energies around several millijoules. Successful desorption has also been observed with less energetic but intense fs-laser beams [96]. The exact mechanism underlying LIAD is still being debated [97]. One explanation is that the laser-induced shockwave breaks the surface-adsorbate bonds. Another attributes the bond breakage to the fast heating of the target surface. Recent experiments [98] have shown that a mixture of these two mechanisms may occur. LIAD creates very slow neutral particles, with a velocity of only a few m/s for particles in excess of 10,000 amu [99]. However, the resulting beam intensities are significantly lower than using MALDI.

In contrast to that, Surface-Assisted Laser Desorption/Ionization (SALDI), uses fast heating of the substrate's front surface. This has been demonstrated in various studies [100-102] always with the goal of breaking the surface-adsorbate bond before a significant amount of energy can be transferred to fragmentation channels of the molecule.

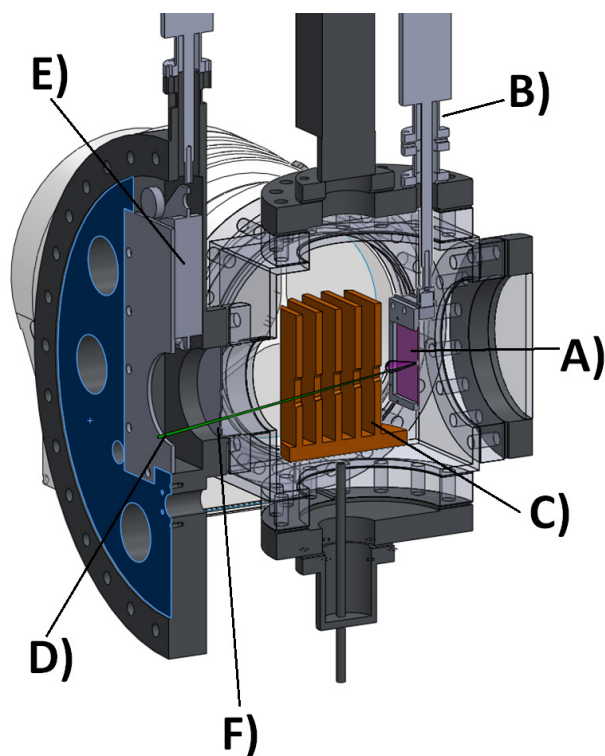


Figure 4.1.3.1: Laser source assembly: A) target plate, B) one of two translation stages, C) liquid nitrogen cooled cryopump, D) laser beam, E) valve, F) differential pumping aperture.

In order to explore the evaporation of large molecules we have built a laser desorption source. The target plate is attached to two computer controlled translation stages inside the vacuum chamber. We use a laser operated at 100 Hz in order to create a pulsed molecular beam. Alternatively, we could chop the molecular beam itself. As the interferometer is pulsed, operating at up to 250Hz, modulating the source saves material and allows us to run the experiment for a longer period of time with the available sample area of 25 cm². It also avoids unnecessary heating of the vacuum chamber walls. This keeps the base pressure low and the molecular transmission to the detector high. During operation we reach a background vacuum of 5×10^{-6} mbar in the desorption chamber. Therefore, only a single differential pumping stage is needed between the source and the interferometer chamber, which is operated at 1×10^{-8} mbar and below. This short setup also minimizes the signal loss which scales, for pulsed sources, with $1/L^3$, where $L = 0.4$

m is the distance between the source and the detector. The additional factor $1/L$, in addition to the expected $1/L^2$ related to the transverse geometrical expansion, is due to the fact that the particle packet stretches in the forward direction as the particles of different velocities separate in space.

The target plate can be coated with particles by drying molecules from a solution at room temperature, or by pressing a molecular powder onto the target plate. Thermally stable molecules can also be evaporated and deposited onto the target substrate.

The desorption laser beam is focused to ~ 0.02 mm. We can use either a natively pulsed laser like an Nd:YAG pumped OPO⁷ (optical parametric oscillator) with nanosecond short pulses and a few mJ/pulse energy, or a 10 W continuous wave (cw) laser⁸ which is mechanically chopped to a pulse length of ~ 20 μ s. The relatively slow heating of the molecules by a chopped cw laser can result in a lower desorption temperature: as soon as the bonds to the substrate are broken, the molecules fly off. Energy that may still be absorbed by the molecule in free flight will only add to their internal temperature but not to their velocity. The drawback of continuous heating is that the particles are more prone to fragmentation than in nanosecond desorption. This can be partially mitigated by illuminating the particles from the back side through a transparent sample plate with a short focused laser [11], or by front side illumination at a steep angle. In such arrangements the particles leave the region of high laser intensity very quickly and are therefore not subject to unnecessary heating. Back side desorption of thin coatings, of a few thousand monolayers thickness, yields $10^{22}/2\pi$ molecules from an area of 2 μm^2 [103]. Using substantially thicker samples of a few μm thickness requires front side illumination. Otherwise, material flakes can be blasted off the surface due to the evaporation of buried molecular layers while the top layers are still solid.

Recently, heavy molecules around 22,000 amu tailored to enhance their thermal volatilization, have been observed to have a velocity distribution centered around 45 m/s [104], when being desorbed with a laser from a metal surface. This is compatible with our current interferometer dimensions as it can be seen in **figure 4.1.1**. Hence this source is a good candidate to attempt to set a new mass record in matter-wave interferometry.

⁷ Ekspla NT242-HE-100-SH/SFG

⁸ Coherent Verdi V10

4.1.4. Gas Beam Molecular Source (GBMS)

There are two classes of interesting particles for interference experiments in OTIMA: heavy particles to probe the mass limits of quantum physics and complex particles on which functional measurements [105-107] can be performed. Complex particles like biomolecules are usually not thermally stable. Thus, the use of the already described sources is not a viable option.

In order to bring complex biomolecules into the gas phase we have combined a cryogenic Even-Lavie valve with a Surface-Assisted Laser Desorption setup [108]. A 1064 nm laser is used because most biomolecules are transparent at IR wavelengths [109]. This ensures that their internal degrees of freedom are only minimally heated [108], enabling most of the energy to be deposited into the substrate. The substrate surface is heated at a rate of 10^8 - 10^{13} K/s [110] for nanosecond and picosecond pulsed lasers, respectively. The superheated surface transfers some of its energy to the adsorbed particles while the rest diffuses into the bulk material. At heating rates above 10^{10} K/s it has been observed [110, 111] that most of the energy goes into the surface bonds and not the internal degrees of freedom of the molecules. It has been proposed [112, 113] that the energy from the hot surface is transferred to the molecules preferentially through surface-adsorbate bonds. These are typically van-der-Waals bonds which have low-frequency vibrational modes, similar to the surface phonons of the substrate [108]. Since the internal vibrational modes of the molecules are at higher frequencies this mismatch is supposed to limit the energy transfer and to create a “bottleneck” for energy flow between the surface bonds and the internal bonds [112]. It is believed that for this reason adsorbate bonds break before the internal molecular degrees of freedom are significantly heated. In order to reduce thermally induced fragmentation after the volatilization process, a supersonic pulse of cold gas is sent over the emerging plume of laser-desorbed particles. These collisions cool the analyte particles and they may also assist in the formation of clusters.

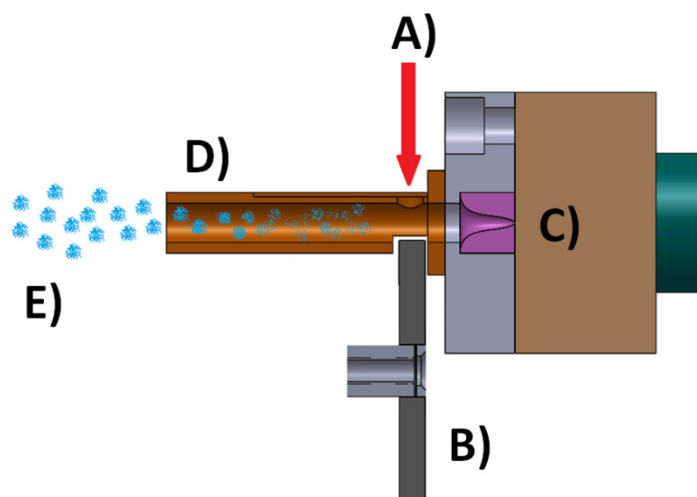


Figure 4.1.4.1: Schematic depiction of the GBMS core elements: A) laser pulse, B) rotating glassy carbon wheel, C) cryogenic pulse valve, D) thermalization tube, E) particles. The thermalization tube is in good thermal contact with the valve which is cooled with a cryostat.

We use a nanosecond pulsed Nd:YAG laser⁹ with a wavelength of 1064 nm and a pulse energy of up to 100 mJ, giving us surface heating rates in the range of 10^{11} K, to desorb the molecules. The substrate is made from glassy carbon which has high absorptivity in the infrared and which can withstand high laser powers [114, 115]. The glassy carbon is machined into a 30 mm diameter wheel. This rotates tangentially to the collision channel and is positioned immediately after the nozzle of the Even-Lavie valve. This way it continuously introduces a fresh surface to the desorption laser. It is driven via a vee-belt by a motor¹⁰ with a 173:1 gear. The desorption wheel is pressed against a counter-rotating brush wheel that is driven with another vee-belt from the main shaft. This brush wheel is made of felt and is used to pick up fresh molecules from a slide underneath to deposit a thin layer of molecules on to the carbon wheel. The supply slide is moved slowly with a translation stage and can be filled with up to 100 mg of molecules, which is sufficient for a few hours of signal. Both the thermalization tube and the valve are in good thermal contact and are cooled with a sterling cooler¹¹ to 80 K.

⁹ Innolas Spitlight 400

¹⁰ Faulhaber Minimotor SA 22/2K 173:1

¹¹ Ricor - Stirling Cooling Technology

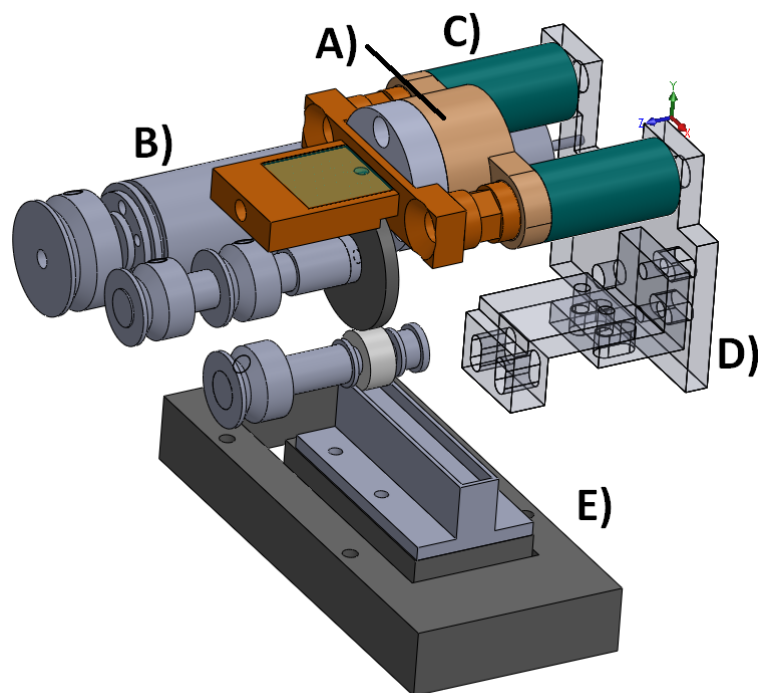


Figure 4.1.4.2: Schematic depiction of the full GBMS assembly: A) The core assembly see **figure 4.1.4.1**, B) driving motor, C) thermal isolation, D) source mount plate, E) supply slide and translation stage. The source is picking up particles with a brush wheel from the supply slide and depositing them on a glassy carbon wheel. The particles are then desorbed by a nanosecond pulsed laser and cooled immediately afterwards with a cold gas pulse.

The particles are detected using a TOF-MS, for that they need to be photoionized first. As many large biomolecules are difficult to ionize [116] we chose Gramicidin D, a 1,836 amu heavy polypeptide with 15 amino acids, which is easy to ionize [91], due to its three tryptophan molecules.

4.1.4.1. Source performance and optimization

We have tested different geometries of the thermalization tube to obtain low particle velocities while maintaining high signal intensity. For our default tube we chose a diameter of 4 mm and a length of 30 mm. We observed that a broader collision channel causes a more effusive, slower but less brilliant molecular beam, see **figure 4.1.4.1.1**.

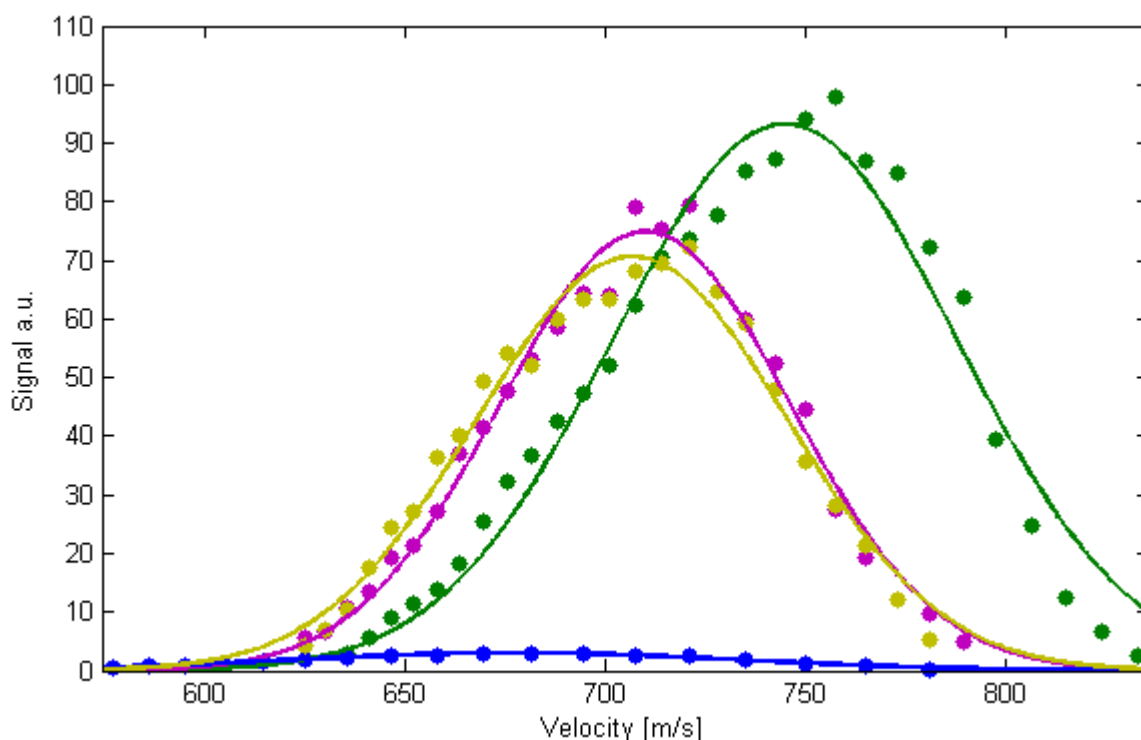


Figure 4.1.4.1.1: Velocity distribution of Gramicidin D in the GBMS Source for different tube geometries at room temperature. The green line represents the data obtained using a 4x30 mm tube; blue line: tube dimension 4x60 mm, yellow line: tube dimension 4x15 mm, and the purple line is a broad tube with tube dimension 8x30 mm.

The tube facilitates cluster formation because of the higher average density over a more extended time – in comparison to a free jet expansion [90, 91]. We have produced pure tryptophan clusters spanning 1-38 molecules, as well as gramicidin monomers, dimers and trimers. In previous experiments, done with a uncooled setup [90], it has been observed that in order to create large tryptophan clusters the addition of CaCO_3 was necessary. In a cooled thermalisation tube tryptophan clusters can be easily created without additives. The respective mass spectrum is shown in **figure 4.1.4.1.2**.

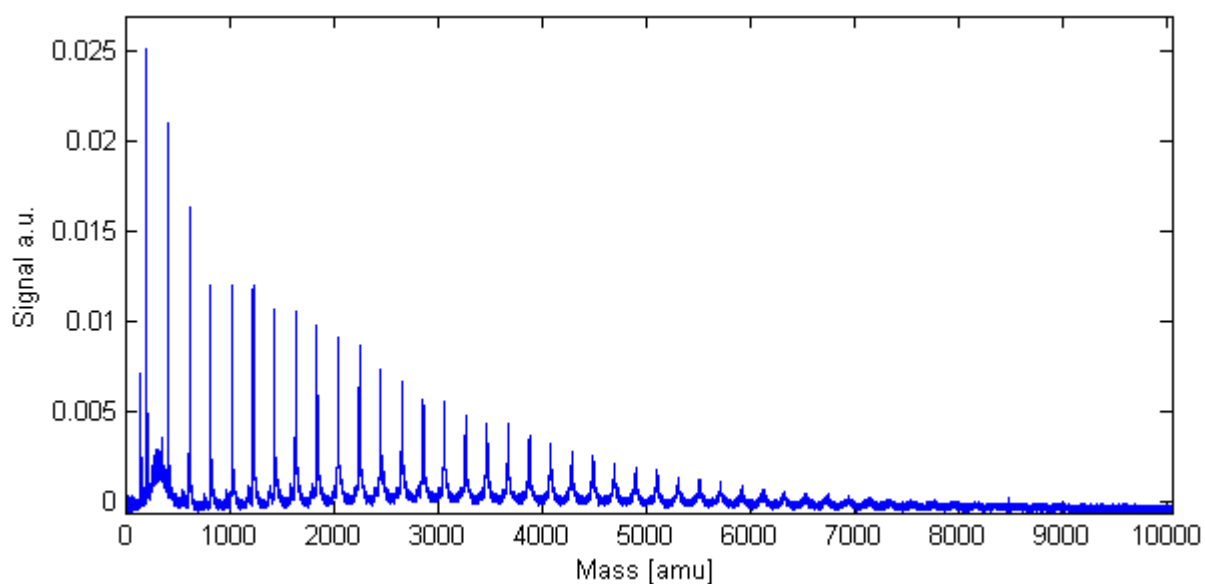


Figure 4.1.4.1.2: Tryptophan cluster mass spectrum with the thermalization tube cooled down to 77 K using 50 bars of neon as buffer gas.

However, the gas confinement in the tube also contributes to the acceleration of the particles. We observed the slowest particles without the thermalisation tube, as shown in **figure 4.1.4.1.3**.

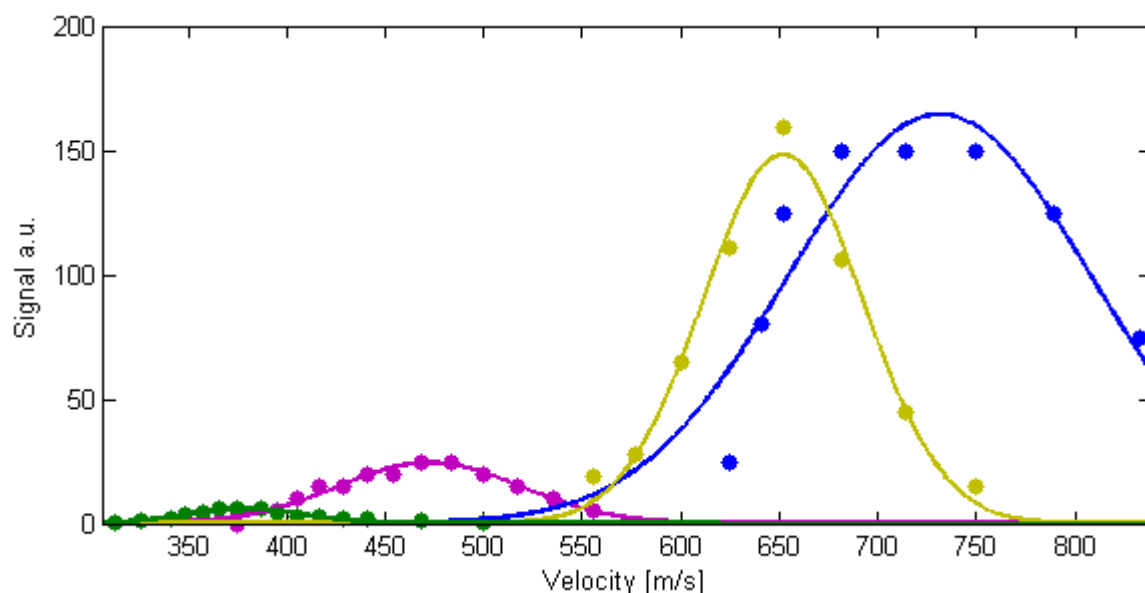


Figure 4.1.4.1.3: Velocity distribution of a single Gramicidin D in the GBMS Source for different source configurations, using argon as carrier gas. Blue data: gas and thermalisation tube at 300 K; Yellow data: without the thermalisation tube, gas at 300 K; Purple data: with gas and tube at 102 K; Green data: without the tube, gas at 96 K.

With the current interferometer geometry one can expect to observe interference in the first Talbot order for particles with a mass up to $\sim 4,000$ amu. Using Helium with a temperature of 4K as a carrier gas one might increase this limit to $\sim 8,000$ amu. This source enables the OTIMA apparatus to do interference experiments with medium sized complex biomolecules. Possible new candidates could be, for example, indolicidin, polytryptophan, or other polypeptides that can be ionized in our TOF-MS.

4.2. Vacuum setup

Environmental decoherence is an important issue in matter-wave interference experiments [25, 61, 117, 118], with two processes being dominant. One is the collision of the delocalized particles with residual gas molecules inside the vacuum chamber. The other is the emission and absorption of thermal photons while the particles pass through the interferometer.

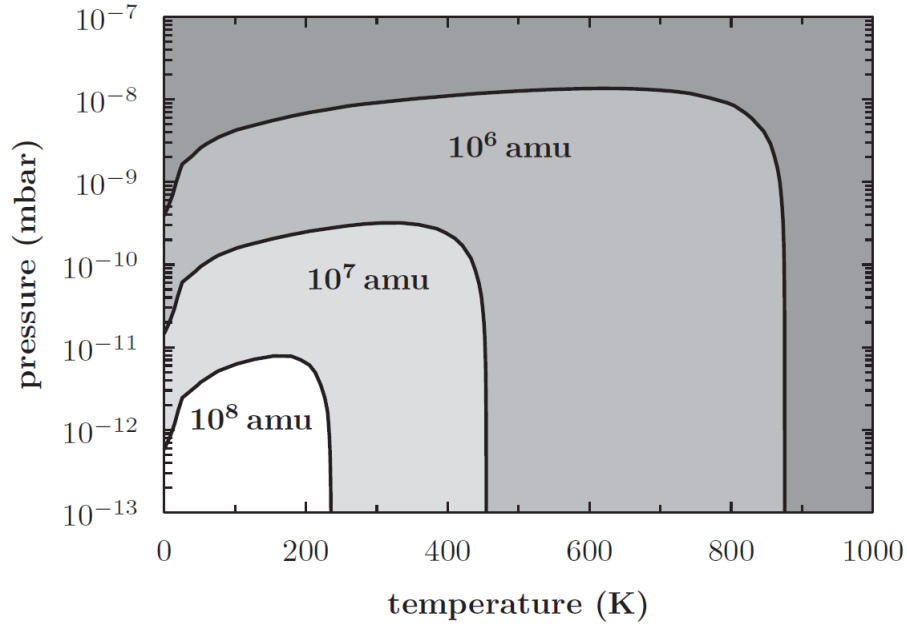


Figure 4.2.1: Simulation of the background pressure and cluster temperature that will reduce the fringe visibility by a factor of two in OTIMA interferometry, in a N_2 atmosphere. The data and the image are computed for gold clusters and taken from [55]

The collision cross section and the optical absorption cross section both grow with the particle size. Hence the constraints on the environmental conditions tighten with increasing particle size. Estimates using the current OTIMA [55] configuration suggest that interference of 10^6 amu large gold clusters can still be achieved at room temperature with a background pressure of 10^{-9} mbar. For particles as produced in the already presented sources, ranging up to 10^5 amu, a residual gas pressure below 1×10^{-8} mbar is sufficient.

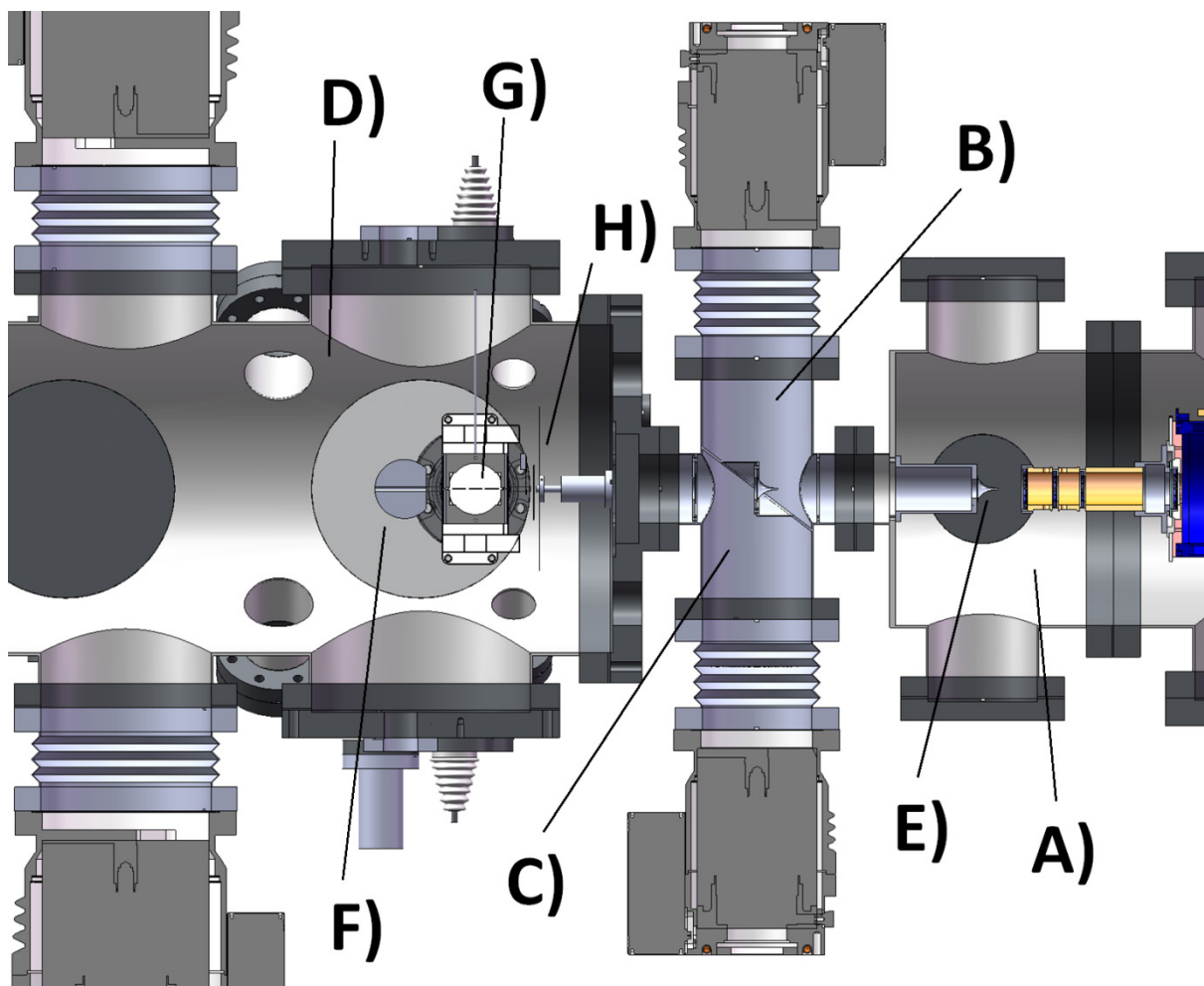


Figure 4.2.2: Complete vacuum setup; cross section through the OTIMA apparatus: A) source chamber here with the magnetron sputter source (various other sources have been used as well, as described in the previous chapter), B) first differential pumping stage, C) second differential pumping stage, D) main experiment chamber, E) first skimmer, F) TOF-MS, G) interferometer assembly, H) chopper disk.

The OTIMA apparatus can currently reach a vacuum of 2×10^{-9} mbar in the main chamber. We use three turbo molecular pumps¹² with a total pumping speed of 1700 l/s. Due to flow restrictions imposed by the vibration damping bellows the effective pumping speed is limited to about 1000 l/s. When using a source with a significant gas load two differential pumping stages with skimmers [15] are inserted between the source and the main chamber. The setup as it is cannot be baked out at temperatures above 100°C due to the fragile CaF_2 windows, required for the grating lasers. State of the art vacuum chambers have been reported to reach 10^{-17} mbar when cooled to 4.2 K [119].

¹² Pfeiffer Vacuum HiPace series

4.3. Beam paths and lasers

The interferometer uses three VUV (Vacuum Ultra Violet) fluorine excimer lasers¹³. The fluorine molecule has two predominant emission lines [120] with a width of $\Delta\lambda \sim 1$ pm. The stronger one of the two has a wavelength of 157.63 nm and contains about 90 % of the total intensity. The second line has a wavelength of 157.52 nm and contains the remaining 10 %. We don't filter out this line as no filters are available at this wavelength which would remove it while allowing the main line to pass. Using a CaF₂ Fabry-Perot interferometer, we have estimated the longitudinal coherence length of these lasers to be ~ 14 mm and we estimated the transverse coherence, based on the laser aperture, to be 50-100 μm [56]. The laser profile is approximately a flat top 2×9 mm in size. Each laser has a maximum pulse energy of ~ 5 mJ, with a stability of about 2% under optimal conditions and better than 10% otherwise. Key factors which affect the stability are the fluorine pressure inside the laser and the power used to pump the gain medium. The lasers have a pulse length of 7-9 ns and a temporal jitter of about 4 ns. There is also a long term drift of a few hundred nanoseconds that is monitored in real time and compensated by software, see the **chapter 4.6.2** for more details.

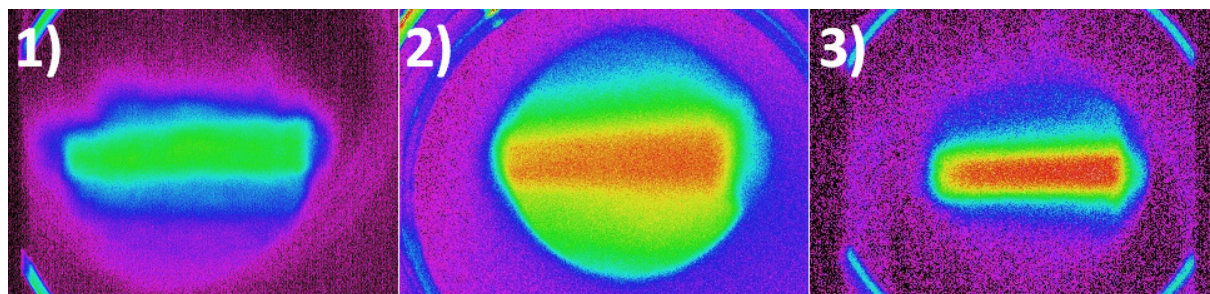


Figure 4.3.1: We have inspected the laser profiles at the laser exit using a fluorescence plate and a CCD beam profiler¹⁴. The lasers have a roughly rectangular flat top profile. 1) Preparation laser, 2) Diffraction laser, 3) Masking laser.

Light with a wavelength of 157 nm is strongly absorbed by oxygen [121], resulting in a $1/e$ penetration depth of only 40 μm in air. Therefore, all beam paths must either be evacuated to a pressure of better than 10^{-3} mbar, or purged with a clean non-absorptive gas like nitrogen. It has been observed [122] that under vacuum conditions residual traces of hydrocarbons, such as often found in pump oils and grease, undergo photo-deposition and contaminate the optical surfaces.

¹³ GAM LASER – Ex50

¹⁴ Ophir-Spiricon SPCAM SP620U 1/1.8" CCD 1600x1200

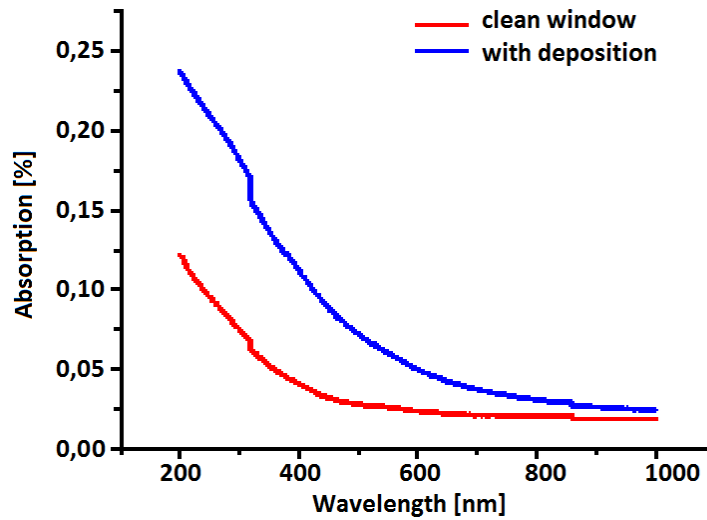


Figure 4.3.2: We examined a contaminated CaF₂ window using a UV spectrometer at the *Institut für Quantenoptik und Quanteninformation* (IQOQI). We have observed a significant degradation of transmissivity at 200 nm. The spectrometer couldn't measure at 157 nm, but the effect is expected to be more severe at shorter wavelengths.

Purging the system typically prevents this from happening. In our setup the beam paths are evacuated below 10^{-5} mbar when the machine is idle and purged with 99.9999% pure nitrogen at 1 mbar during operation. These measures showed to be very successful in preventing optics degradation. However, the main experiment chamber must be kept at high vacuum, without purging, and the window to the chamber as well as the interferometer mirror degrade with time. This contaminants can be largely removed by purging the main chamber, using a purge gas with trace amounts of oxygen or water added, while the laser is operating [123].

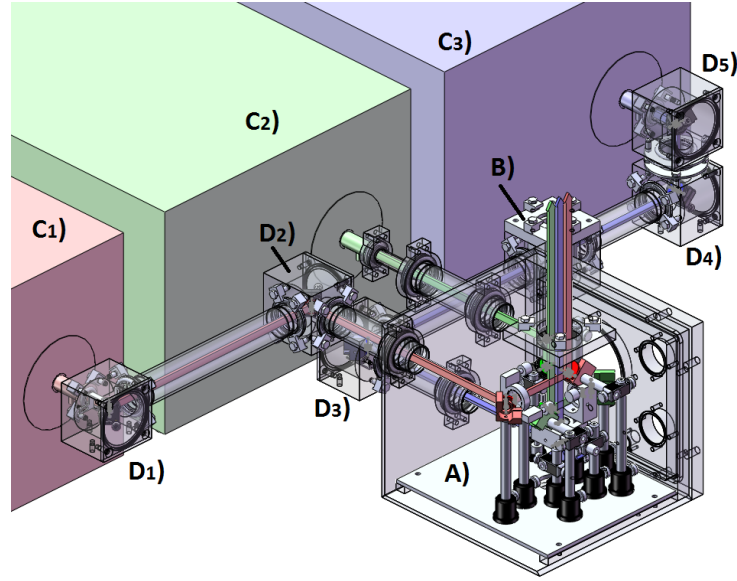


Figure 4.3.3: VUV beam path: the beams of all three lasers ($C_{1,2,3}$) are guided by mirrors inside the vacuum tubes ($D_{1,2,3,4,5}$) to the main box (A), where each beam is focused by a cylindrical lens and guided with mirrors upwards through a CaF_2 window into the interferometer chamber and onto the interferometer mirror. The middle grating laser goes additionally through an absorption cell (B). The three lasers and their paths are color coded for better visualization.

The beam line comprises a main box and three smaller boxes containing one mirror each. They are all connected with tubes. Each beam path contains 4 mirrors, two of which are computer controlled. This allows the interferometer beams to be adjusted while the system is closed. The laser beams are guided into the main optic box, each through a cylindrical lens with a focal length of $f \approx 60$ cm. They are lined up in a row and directed upwards onto the interferometer mirror. The beam line system is separated from the interferometer chamber using a CaF_2 window. In addition, the middle laser beam propagates through an absorption cell which enables precise control of the optical power entering the interferometer. By controlling the oxygen pressure inside the cell between 10^{-4} mbar and 200 mbar, we can tune the laser power from 100% down to almost 0%. Photodiodes¹⁵ are used to measure the pulse energy of each beam in-situ and in real time. This allows us to post-select the acquired data according to both timing and energy criteria and enables feedback where necessary. The main box is also equipped with a pyroelectric sensor¹⁶, which can be moved into the path of each laser to enable calibration.

¹⁵ Thorlabs GaP Photodiode FGAP71

¹⁶ Ophir Pyroelectric Laser Energy Sensors PE25-C

4.4. Interferometer assembly

The core part of the OTIMA apparatus is the interferometer assembly. It harbors a single CaF_2 mirror with a diameter of 50 mm on which three laser beams are retro reflected to create standing light waves. These act as absorptive gratings which form the interferometer. Using a single mirror for all three lasers simplifies the beam alignment considerably. The mirror provides a common boundary condition for the lasers thereby defining the direction of the standing light waves. The grating period is $d = \lambda/2 = 78.8 \text{ nm}$ at 0° incident angle, where $\lambda = 157.6 \text{ nm}$. However, when the laser beam is tilted by an angle θ the modulus of the wave vector perpendicular to the mirror is reduced to $k' = k \cos \theta$, thus stretching the period of the light grating. Hence, the grating period of each laser is given by:

$$d = \frac{\lambda}{2 \cos \theta}$$

The whole adjustment of the interferometer can be done by retro reflecting the three lasers while ensuring they fully overlap with the particle beam.

A common problem with any interferometry setup are vibrations, here of the wavefront-defining mirror surface, which are a major source of dephasing [124]. They can wash out the interference patterns by introducing time dependent grating shifts. If these are greater than half the grating period the interference contrast will be washed out completely.

The single mirror architecture makes OTIMA less sensitive to vibrations and drifts. When the single mirror tilts all three gratings are tilted identically, and their effective periods change the same way. The displacement of the interference pattern relative to the third laser grating is given by [44]:

$$\Delta_{rel} = \Delta_1 - 2\Delta_2 + \Delta_3$$

where $\Delta_1, \Delta_2, \Delta_3$ are the shifts of the individual laser gratings. Due to the intercept theorem a shift at the second grating displaces the interference fringes twice as much than at the first or third grating. Thus, the observable displacement Δ_{rel} remains unaffected. As long as this happens on time scales much longer than the particles flight time through the interferometer, the interference patterns will not wash out.

High frequency vibrations, however, are still a problem as they will change the observed phase randomly and thus average out the interference patterns. This can be avoided by measuring the mirror movements while the particles are passing through the interferometer and averaging the measurements into separate bins based on the measured phase shift. A similar method is used in atom interferometry [125].

The mirrors used in OTIMA were manufactured by Jenoptik and have a reflectivity of 96% at a 0° angle of incidence. This was the best available standard at the VUV wavelength of 157 nm [123] at the time of purchase. Some of the mirrors were examined by Laser Optik Garbsen, by the Physikalisch-Technische Bundesanstalt in Germany and by Bruker. Their measurements showed that the surface has a roughness of 60 nm (variation from peak to valley) over the whole clear aperture. Imaging of selected spots on one of the mirrors done at the TU Wien with an AFM (Atomic Force Microscope) showed the local surface roughness to be around 10 nm.

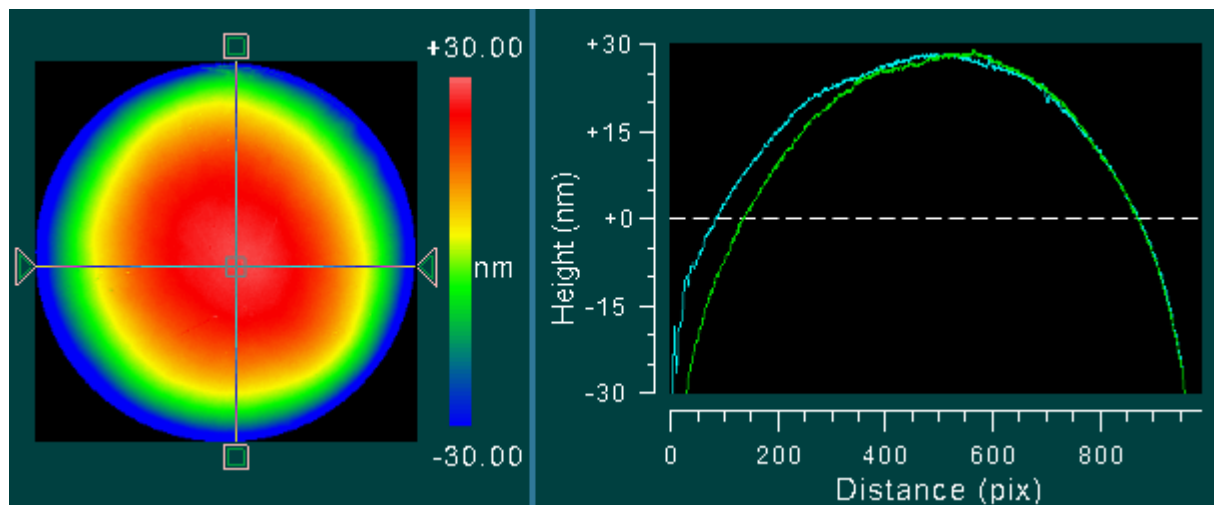


Figure 4.4.1: Interferometer mirror surface height as measured interferometrically at 632 nm by PTB Germany.

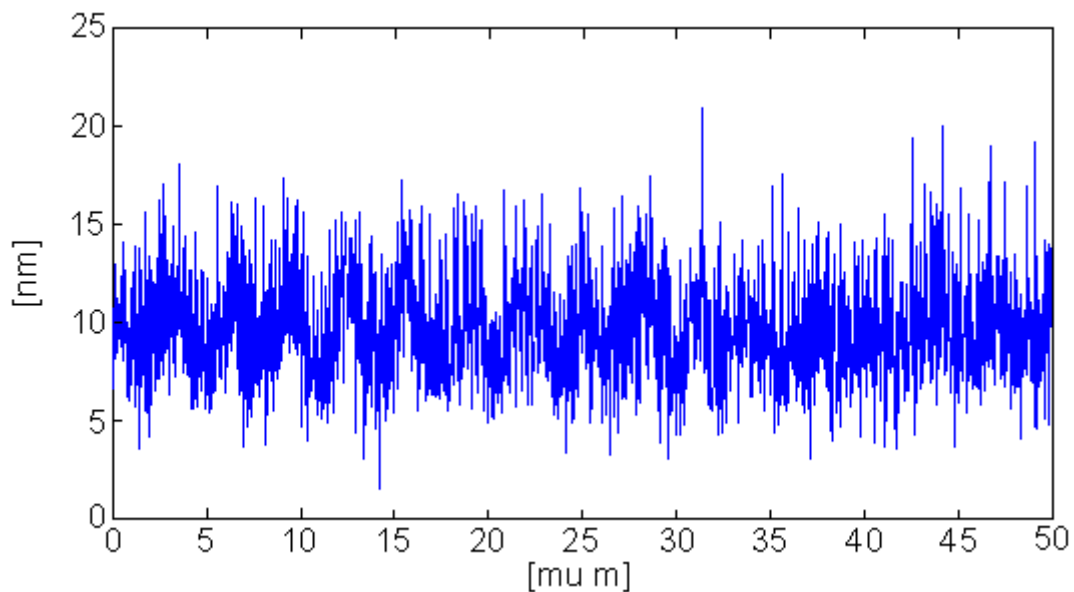


Figure 4.4.2: Cross section through a $50 \times 50 \mu\text{m}$ AFM image of the interferometer mirror surface measured at the TU Wien.

The small scale surface features don't pose a problem but the convex deformation over the entire mirror is disadvantageous. It introduces a displacement between the grating planes as well as an angle. With the measured mirror curvature the first and third grating are tilted by $0.2 \mu\text{rad}$ with regard to the middle grating. The angle causes a phase shift within each grating itself. Thus, particles which start at different positions along the beam direction will experience different grating phases, like it is illustrated in **figure 4.4.3**.

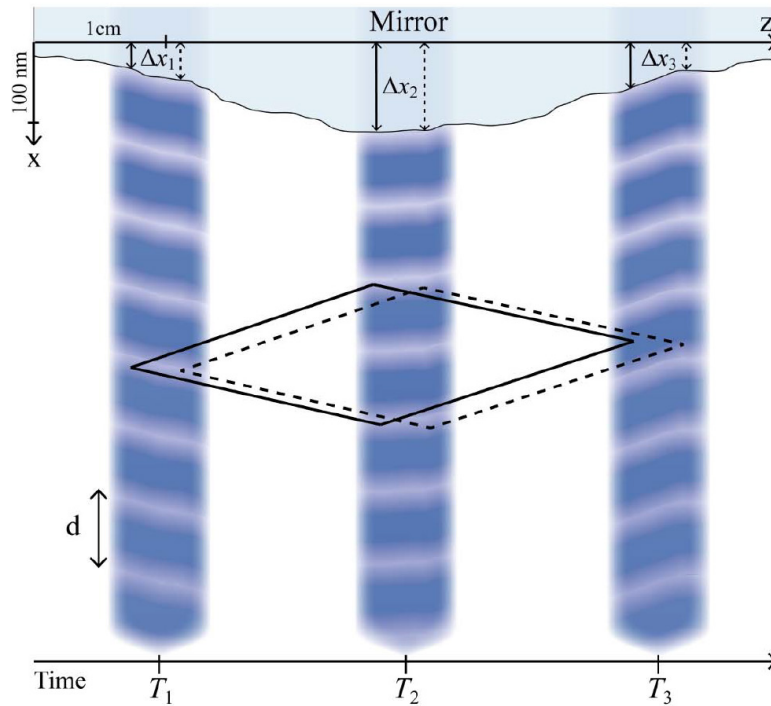


Figure 4.4.3: Interferometer mirror with three standing laser light waves. The figure is not drawn to scale, in order to illustrate the effects the surface irregularities have on the light gratings. The solid and dashed lines represent semiclassical paths of two different particles through the interferometer, for each $\Delta x_{1,2,3}$ marks their respective grating shifts. In this image one particle ends up in the antinode of the third grating while the other passes through an antinode, so interference contrast is smeared out. The figure is taken from our publication [56].

To mitigate this effect the particle pulse can be cropped, by restricting the ionization laser in the detector, such that for example only 5 mm along the beam direction is used. In this particular case the phase shift between the begin and the end of the selected section would be about 10 nm, resulting in a worst case visibility reduction of 13%. If the tilt angle were $1.6 \mu\text{rad}$ the phase shift over the same 5 mm would be half the grating period thus completely smearing out the interference pattern. In everyday use the particle beam is usually cropped to 1 mm length.

The phase shift introduced by the constant displacement of the grating planes can also reduce the interference contrast. By varying the laser pulse timing different spots of the mirror can be addressed. This way this effect can be partially compensated.

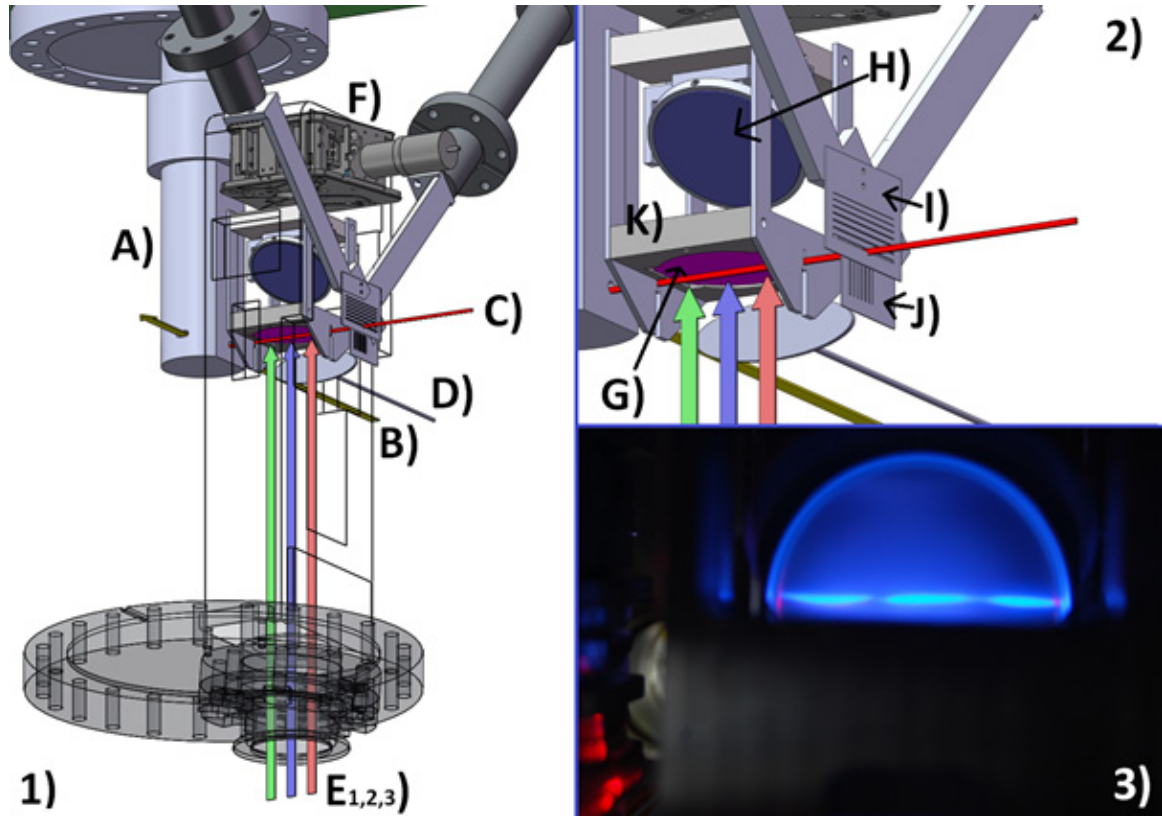


Figure 4.4.4: Interferometer assembly 1) consists of: A) TOF-MS, B) ionisation laser, C) particle beam, D) deflection electrode, E_{1,2,3}) three VUV grating laser beams, color coded for better visualisation, F) linear stage allowing to lift the mirror. The mirror cage 2) contains: G) interferometer mirror, H) auxiliary UV enhanced aluminum mirror, I) horizontal delimiter slits, J) vertical delimiter slits, K) mirror holder block. 3) View into the chamber while the grating lasers are operating

The interferometer assembly is mounted on the flange interfacing with the main optic box described in the previous chapter. In front of the assembly there are two adjustable slits, one limiting the width and the other the height of the particle beam. This way it can be ensured that the entire beam which enters the interferometer will fully overlap with all three lasers. And that it will pass close enough to the mirror surface to see a good standing light wave despite the short longitudinal coherence of the lasers. The interferometer mirror rests on its circumference on a 1.5 mm large brim inside the mirror holder block. This way the forces acting on the mirror are minimal and its deformation under the influence of gravity is minimized, see **figure 4.4.5**.

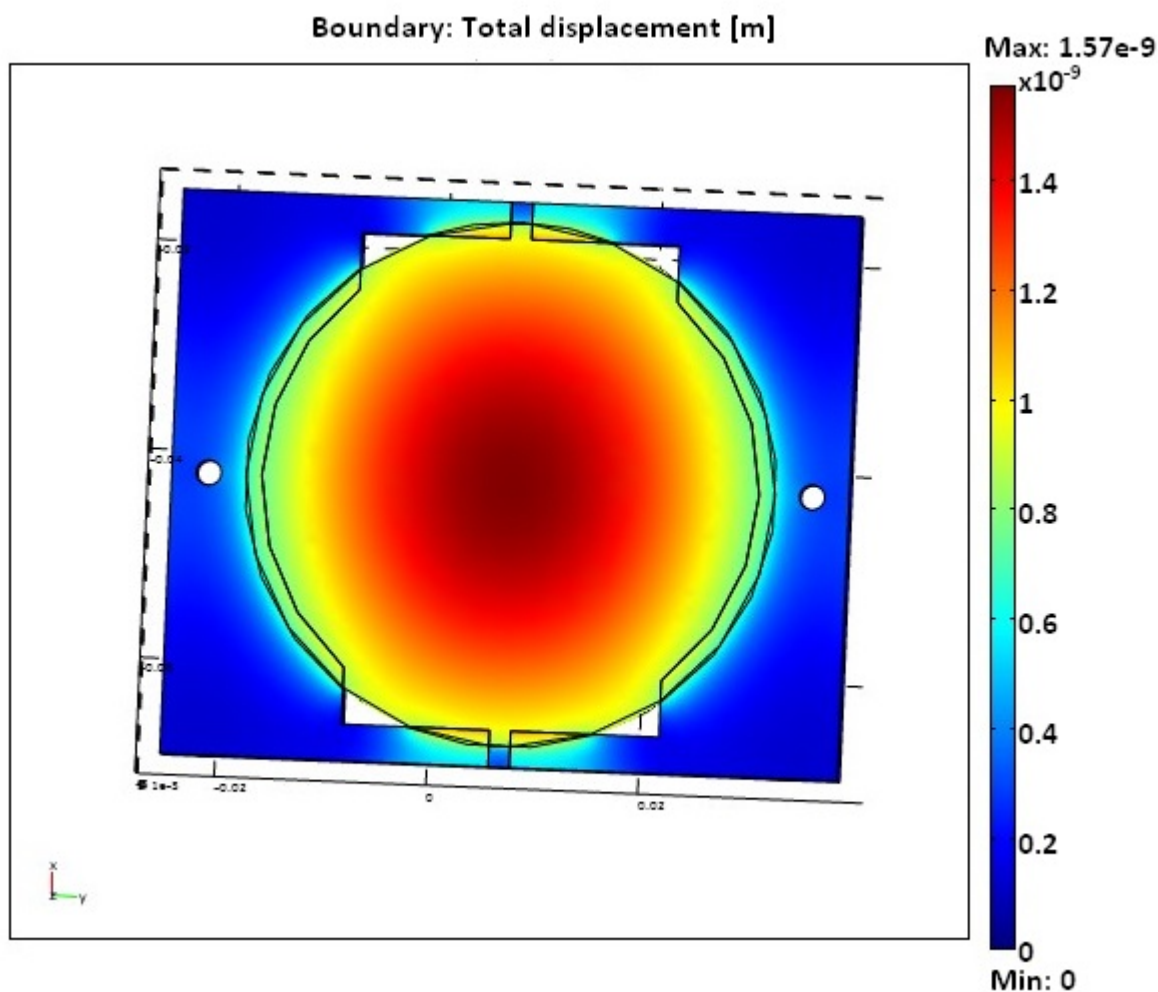


Figure 4.4.5: COMSOL¹⁷ simulation of the CaF₂ mirror deformation under gravity shows a deformation of only ~0.8 nm in the mirrors clear aperture.

The mirror cage is mounted on a translation stage¹⁸ allowing to move the mirror up and down by up to 13 mm. This way the mirror can be lifted sufficiently high to ensure that the particles will see only running waves and no longer standing light waves from the grating lasers. This can be used to investigate the beam depletion mechanism and particle properties. Above the main CaF₂ mirror we mounted a UV enhanced aluminum mirror which allows observing the fluorescence on the main mirror as well as introducing additional laser beams if needed. Once the particles passed through the interferometer they enter a time-of-flight mass spectrometer where they are ionized with another VUV laser and detected.

¹⁷ COMSOL Multiphysics® Modeling Software

¹⁸ MICOS Stage

4.5. Detector (TOF-MS)

The OTIMA interferometer has a pulsed mode of operation and the created interference patterns depend highly on the particle mass. Therefore, a time-of-flight mass spectrometer (TOF-MS) was selected as a suitable detector. The mass spectrometer was custom made for the experiment¹⁹. It has a mass resolution of $\Delta m/m = 1:5000$. This type of mass-spectrometer operates by accelerating ions in the detection region with a strong electrical pulse and measuring the time the ions need to reach a detector at the end of a drift tube. The ideal flight time of the ions is equal to

$$t_{of} \approx \sqrt{\frac{m}{z}}$$

where m is the ion mass and z its charge. However, different starting positions of the ions in the detection region as well as a spread in their starting velocities can diminish the mass resolution. In our instrument and for typical parameter settings, the mass calibration is $m [amu] \approx t^2[s] * 1.2 \times 10^{12}$ for a time of flight t given in seconds. The day to day reproducibility is about 0.05%, mostly limited by the resolution of the voltage displays.

To compensate for the different starting positions a 2-stage acceleration scheme is used as described by Wiley and McLaren [126]: ions starting lower in the detection region will be accelerated more than ions starting closer to the 2nd stage. If the voltages are selected correctly, all ions catch-up to reach the detector at the same time after the drift tube. To compensate additionally for different starting velocities, a dual-stage ion reflector [14] is used: faster ions penetrate deeper into the reflector assembly and are delayed more than slower ions. With the right configuration this allows the faster ions to fall behind just enough that after a second drift tube all ions of the same mass reach the detector at the same time.

At the end of the second drift tube the ions are post accelerated with up to -3 kV onto a triple Multi-Channel-Plate (MCP) stack (Z-Stack configuration [127]). Together with the voltages of the drift tubes (liner 1 and liner 2) they reach an ion energy up to -20 kV, which is necessary to efficiently eject secondary electrons in the MCP for larger ions. This is due to the detection efficiency of a MCP scaling with the ion velocity and not its kinetic energy. Thus, the detection efficiency decreases with increasing particle mass [128].

¹⁹ Stefan Kaesdorf, Geräte für Forschung und Industrie, München

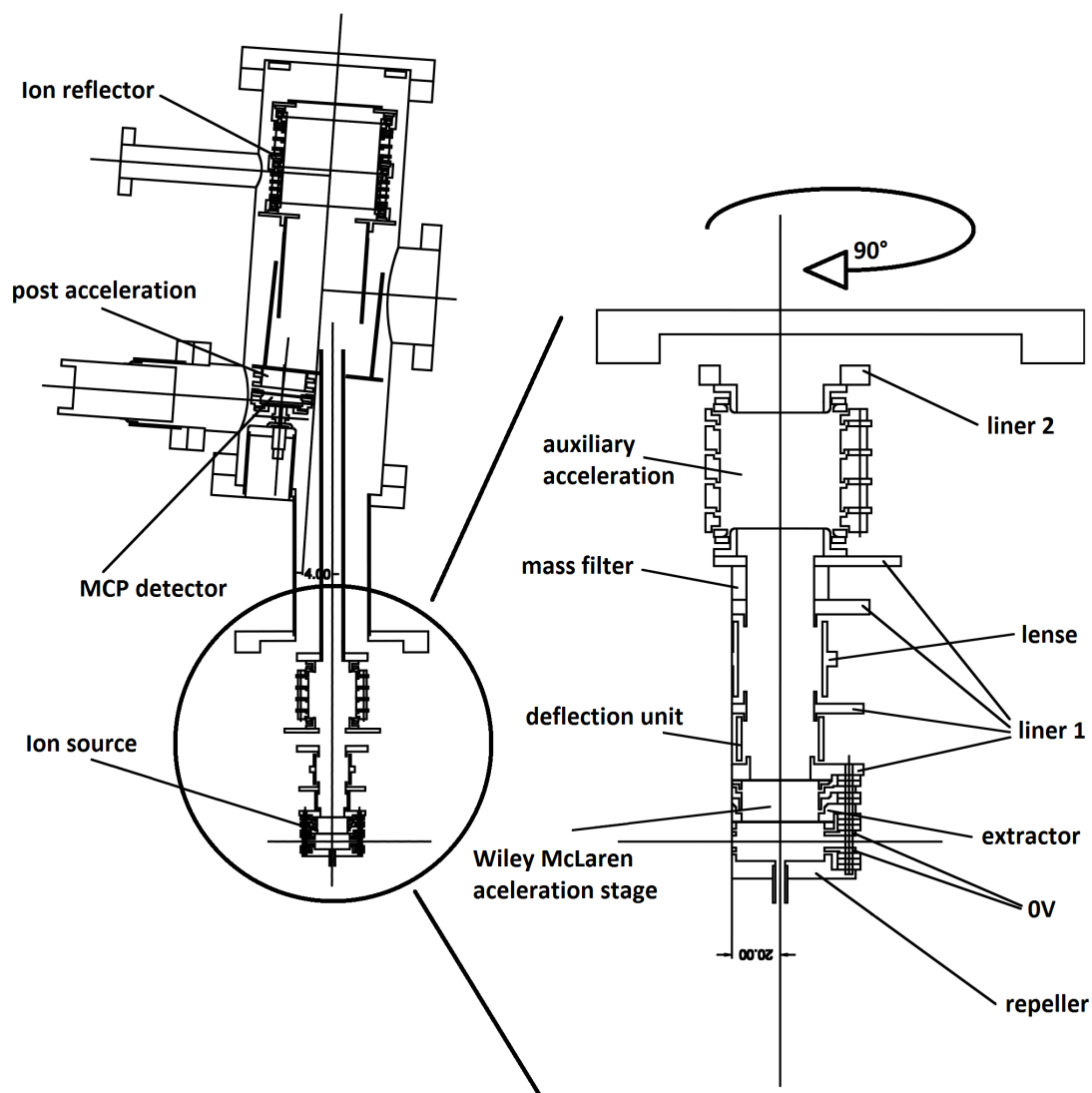


Figure 4.5.1: Schematic depiction of the reflectron type TOF-MS used in the OTIMA setup. The ions are accelerated in a Wiley McLaren arrangement. In the first stage two opposite voltages are applied to the repeller and extractor which give the ions their initial acceleration. In the second stage the ions are accelerated further with the voltage of liner 1. Then the ions move through the first drift tube. The first drift tube contains additional ion optics which allows guiding the ion beam. Then they are accelerated further to the liner 2 potential and enter the second drift tube. This auxiliary acceleration is necessary to reach the final TOF voltage of up to 16 kV, implementing all the guiding optics directly at the full voltage would be very expensive. At the end of the second drift tube the ions are reflected with a dual stage reflector under a 4° angle down to the detector. The detector is a triple channel MCP floated at a potential of up to 3 kV relative to liner 2. Image taken from [129].

The particles that pass through the *Interferometer Assembly* are neutral and need to be ionized in the mass spectrometer. This is done with a fourth VUV Excimer laser²⁰ similar to those used in the interferometer. An issue that arises during the photoionization is that the TOF-MS measures only the mass-to-charge ratio and not the mass directly. Thus if a particle is doubly charged it will appear at the same time as a singly charged particle of half the mass. This can mix-up the mass spectra in cluster beams. This issue can be mitigated by selecting the right ionization energy and wave length to maximize the probability of creating singly charged ions. Alternatively if the mass distribution of the clusters is sufficiently large and uniform, the multiply charged signal can be extracted for ions where $m/(z \cdot e)$ is not a multiple of $m/(1 \cdot e)$, then approximated for all multiply charged ions and finally subtracted from the mixed signal. This procedure is described in more detail in the **chapter 4.6.3.2**.

²⁰ Coherent ExiStar EX 500

4.6. Measurement and control

The overall design of the OTIMA imposes stringent requirements on the control and data acquisition system. A major set of challenges is posed by the excimer lasers, as described in the **chapter 4.3**. They have a pulse timing jitter of 4 ns and power variations of up to 10%. And even though these are about the best values one can get on the market at the required wavelengths, it makes real time data evaluation and post selection, as well as an online feedback and laser adjustment a necessity. Also, since the cluster transmissivity at each grating is required to be less than about 20%, the total transmissivity of the *Interferometer Assembly* is lower than 0,8%, see **chapter 3.2**. Given the limited brightness of the available sources described in the previous chapters, only a few ions per pulse are expected to come through and reach the detector. Because of pulse to pulse fluctuations as well as long term drifts in the source intensity the signal needs to be averaged to obtain statistical certainty. For that reason a high repetition rate of the experimental cycle is beneficial. The lasers can run at up to 250 Hz which was the goal for the experiment. The need to evaluate each laser pulse individually necessitates the use of a high speed digitizer card²¹. The required record length for each run is the time window from the first laser pulse till the end of the time of flight mass spectrum. For example for a 10-fold anthracene ($178 \text{ g} \cdot \text{mol}^{-1}$) cluster, the Talbot condition sets a time of 54 μs from the first to the last grating laser pulse, plus another $\sim 50 \mu\text{s}$ to reach the detector. Recording the mass spectrum up to $\sim 50,000 \text{ amu}$ requires 200 μs with our TOF-MS. This adds up to a data record length of almost 400 μs , at 10 bit voltage and 1ns time resolution, resulting in 0.8 MB per data record. Running the apparatus at 250 Hz yields a data rate of 200 MB/s that needs to be processed in real time, and respectively more for heavier particles.

This task is handled by our proprietary software MOPS that I also developed within this PhD thesis. It is installed on a dual Xeon workstation with 30 GB of RAM that contains the digitizer card and controls three programmable pulse/delay generators²², as well as other instruments, such as: flow controllers²³, pressure gauges, OPO laser²⁴, motorized optic mounts, National Instruments IO Hardware, motor drivers and other devices.

²¹ Agilent Digitizer U1065A: 10-bit, 8GS/s, 3GHz analog bandwidth

²² BNC Model 575 pulse/delay generator

²³ Aera FC-7700CD

²⁴ Ekspla NT242-HE-100-SH/SFG

4.6.1. Molecular Optics Programming System (MOPS)

The MOPS software was designed to control the experiment and handle hundreds of megabytes of data per second, while providing a simple user interface, allowing for flexible variations of the experiment without the need for an everyday developer intervention. The high demands for the real time data processing made it necessary to use an efficient programming language. The MOPS core application with all its plugins consists of about 75,000 lines of C/C++ code, not counting external libraries. Implementing the software for OTIMA using LabView or MATLAB would not be feasible due to the limitations and *execution overheads* of this more widely used solutions. C/C++, which is compiled directly to *machine code*, has no execution overhead. It allows the developer to implement a custom memory management mechanism. And wherever necessary (for example in bulk memory copying operations) highly optimized *assembler code* can be used.

An OTIMA experiment is executed by binary plugins that are controlled by two scripts and a set of configuration parameters. The *scheduler script* manages the experimental procedure through plugins that control the hardware. The *processor script* determines how the recorded data are being preprocessed by the plugins and how it is stored. A *scheduler script* always starts executing in the entry function `Main`. It is capable of calling plugin functions with arguments, reading/writing plugin parameters and reading/writing configuration flags. These flags are retrieved by plugins that capture records from the external hardware and they are attached to those records. This way the plugins know how the experiment was configured when the particular record was obtained. This implements a simple synchronization mechanism which is important during the subsequent processing of the data. It is necessary as the data processing is done asynchronously in multiple threads. The *processor script* defines how individual records are retrieved and processed. This is done in *sequences*: a record is pulled from a plugin and then passed on for processing to a sequence of other plugins. Each sequence is executed in an independent thread. Plugins can modify a record, drop a record, or create new records that are then pulled for further processing in another *processing sequence*. This highly parallel mechanism allows distributing the computational load over all available processor cores.

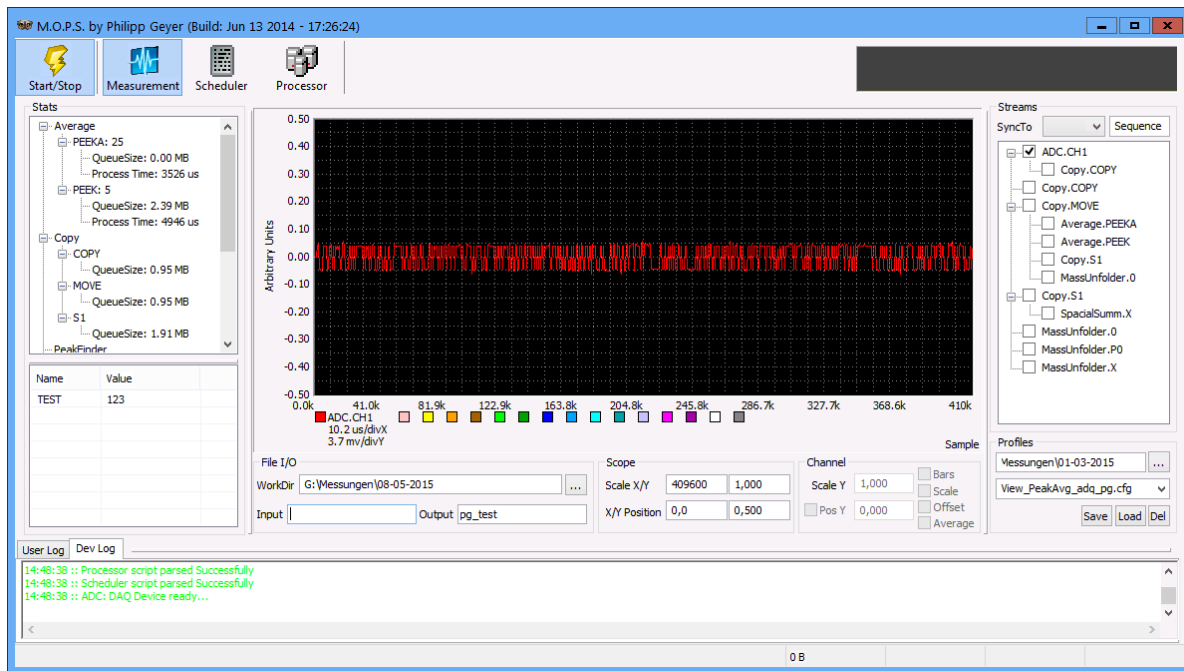


Figure 4.6.1.1: MOPS user interface in the measurement view displaying electronic noise from the digitizer card with no input present. On the left top panel a *tree view* is located, showing real time information provided by the plugins. Below, in the bottom left panel, is a list of current configuration flags. The right panel shows the data processing threads and the processing plugins. Checking one of the items in that tree activates the preview of the current record output. The preview then is displayed in the scope view located in the center. The control elements below the scope view allow setting the data range to be viewed. The panel on the bottom right allows saving and loading configuration profiles for different measurements.

The user interface consists of three views which can be chosen using toolbar buttons:

In the **Measurement view**, currently acquired records as well as all intermediate records can be previewed. It also shows the current configuration flags and plugin information. The *measurement view* offers also the option to manage complete MOPS configuration profiles. A MOPS configuration consists of a list of plugins to be used, their presets, the two scripts, and a configuration for the scope control and the record streams selected for preview. To run an experiment the user must load or create a profile, specify a working directory and an output name where to save data, and press the Start button. If required also an input name can be defined. When a run starts, a plugin can ask for additional parameters or show additional data. During a run plugins and the *scheduler script* can *log events* to a *User-Log*, and/or to a *Dev-Log* (for the software developer). These logs can provide, for example, error information. If parameters need to be changed, or a new profile needs to be created, this is done in the scheduler or processor view respectively.

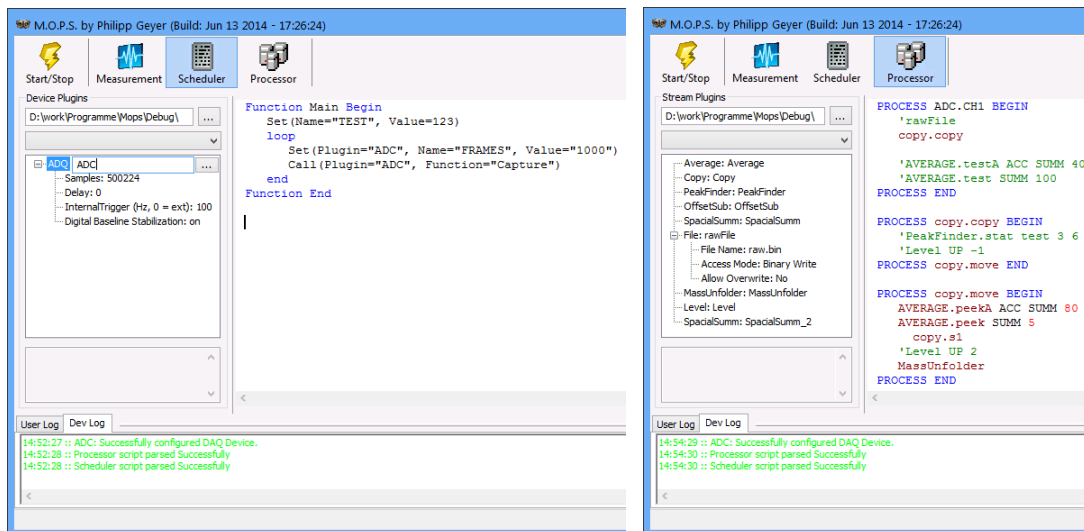


Figure 4.6.1.2: MOPS user interface: the scheduler and processor views show a simple profile used to view averaged digitizer signal. Both views are similarly structured. The left panel allows adding and removing plugins as well as changing their properties. The center panel contains a text editor to modify the respective scripts. The editor provides syntax highlighting functionality.

The ***Scheduler view*** and ***Processor view*** both contain a list of plugins that are to be used. The lists are separated in the user interface for simplicity but handled as one list internally, such that each script can interact with any *instantiated* (that is loaded) plugin. The plugin lists are structured as tree views where the loaded plugin is the root entry and all important plugin presets are shown as children. The tree control is implemented such that important plugin parameters can be entered directly inside the tree view, like for example the sample count that is to be acquired for each record, or the name of a plugin instance. Within the scripts the plugins are always referred to by their unique instance names, hence multiple instances of one plugin can be used. The root entry also presents a button that opens a plugin dialog if the plugin has one. Here, more parameters can be configured and additional plugin information can be viewed.

4.6.1.1. Scheduler script

The *scheduler script*, which was especially developed for MOPS, is a simple *Turing complete* programming language. This means that it supports execution flow-control and can operate on variables. The script is not interpreted from text during execution but is instead compiled into *bytecode* on start which is then executed in a virtual machine. The script engine is implemented in C++ using only the standard type library. Hence, it is very portable and can easily be reused for other projects. See the **appendix 7.1** for further implementation details.

```
Function Main Begin
    FramesToAcquire = 1000 ' how many records are to be acquired

    Set(Plugin="ADC", Name="FRAMES", Value=100) ' set parameter batch size

    Call(Plugin="BNC", Function="ON") ' start experiment

    Batches = FramesToAcquire/100 ' calculate batch count
    loop ((Batch = (Batch + 1)) < Batches)
        Set(Name="BATCH", Value = Batch) ' set a flag
        Call(Plugin="ADC", Function="Capture") ' acquire batch
    End

    Call(Plugin="BNC", Function="OFF") ' Stop experiment

    Get(Plugin="ADC", Name="RATE", Value=Rate) ' read plugin parameter
    LogLine(Line="Acquired data at a rate of: " & Rate & "Hz.") ' report
Function End
```

Code 4.6.1.1.1: An example of a script which sets plugin parameters and calls plugin functions.

In MOPS there are four C++ functions *bound* with the script engine, three of which allow interacting with the plugins. The function `Call` allows executing of a plugin function, it takes the arguments: `Plugin` specifying the plugin name and `Function` with the name of the function to be called. Depending on the plugin interface version used an argument `Arguments`, a string to be passed to the called function, or an arbitrary number of further arguments can be passed to the function. In the latter case all arguments are passed *by reference* to the plugin. The functions called `Set/Get` are used to read and write parameters. They take the following arguments: `Plugin` which is optional, `Name` being the name of the parameter to be accessed and the `Value` to be set. If `Plugin` is not specified the function reads and writes the experiment configuration flags. The fourth function `LogLine` allows writing information to the logs in the user interface.

4.6.1.2. Processor script

The *processor script* is a simple declarative programming language which has also been developed for MOPS. It allows implementing data processing in multiple threads using configurable binary plugins, without requiring the users to take care of all the intricate aspects of *parallel computing*.

A script is made up of processing blocks which represent individual processing threads. Each such thread executes a list of plugin operations: it pulls a record from a plugin, runs it through a list of processing plugin instances and, depending on the script, either deletes the record or returns it to a plugin for further handling. See the **appendix 7.2** for further implementation details.

```
PROCESS ADC.CH1 BEGIN
  COPY.preview
  FILE.raw
PROCESS END

PROCESS COPY.preview BEGIN
  AVERAGE.prevA ACC SUMM 80
  AVERAGE.prev SUMM 10
PROCESS END
```

Code 4.6.1.2.1: Sample of a short MOPS processor script, here data are obtained by the digitizer card from channel 1 then copied inside one thread, and averaged for preview in a parallel thread.

A plugin instance can handle multiple data streams, using either a hard-coded or a dynamic naming scheme. Each invocation of a plugin instance for processing can be customized with an argument line.

The streaming preview of MOPS allows viewing the data records while they are being processed. It captures copies of the processed data a few times per second and displays them on the scope control element in the *measurement view*. As the data processing is parallelized, normally pulled records will not be synchronized to each other. If synchronization is required a configuration flag can be selected in the *measurement view* to synchronize the preview. Then, in each preview only records will be shown which all have the flag set to the same value.

4.6.1.3. MOPS plugins

All hardware interaction and data processing in MOPS is performed by binary plugins that are provided as dynamically linked libraries. The scheduler and processor scripts manage the operations executed by the loaded plugins. A plugin can be a control plugin that accesses hardware or third party software, or it can be a data processing plugin that handles data records. If required, a plugin can also do both, like the "ADC" plugin which controls the digitizer card and creates new data records for processing. All plugins have exactly the same interface. Their functions are defined only by which parts of the interface are implemented. The plugin interface is designed to maximize the binary compatibility, such that plugins can be created using different compilers. The interface uses a *factory function* and an *abstract interface class*, see **appendix 7.3** for a detailed interface specification. To date I have implemented 43 different plugins for MOPS, 12 of which are control/hardware plugins.

Most of the plugins are task specific and the important ones will be explained in depth in the following chapters, but there is also a couple of generic plugins.

The most generic plugin is the "Copy" plugin which allows to copy or move data streams from one processing sequence to another. It maintains internally a record queue for each stream that it processes. When a record is pushed to the plugin it is simply put on the respective queue. When it is processed a copy is put on the queue and the original passes the plugin unmodified. When records are pulled from the "Copy" plugin it returns the entries from the internal queue. If required, they can be filtered by flag values, in which case multiple process sequences need to be provided for each expected flag value.

Another generic plugin is the "Filter" plugin: it takes an argument line specifying criteria for the flag values to be let through. All processed records that don't meet the criteria are dropped.

The "File" plugin allows MOPS to save/load records with the corresponding configuration flags to/from a file. This can be done using either a proprietary binary format where high data rates of hundreds MB/s can be reached or using a simple text format that can easily be parsed by third party applications like MATLAB. When saving data in the text format this is done using either one large file containing all records or each record is saved into a separate file, depending on the plugin configuration.

4.6.2. Experiment control & data acquisition

An experimental run of the OTIMA interferometer is made up of a long series of individual measurements. It is usually performed at a rate of 100 to 250 Hz. The measurements are performed in two modes, the symmetric (interference) mode and the asymmetric (reference) mode. In the symmetric mode the grating laser timing is set such that the temporal Talbot-Lau condition is fulfilled and interference contrast is expected to be observed. In the asymmetric mode, however, the timing of one laser is detuned such that no quantum mechanical contrast is expected. These measurements provide a signal baseline to which the interference shots are compared to. These two modes are interleaved in order to compensate for short term fluctuations of the source intensity.

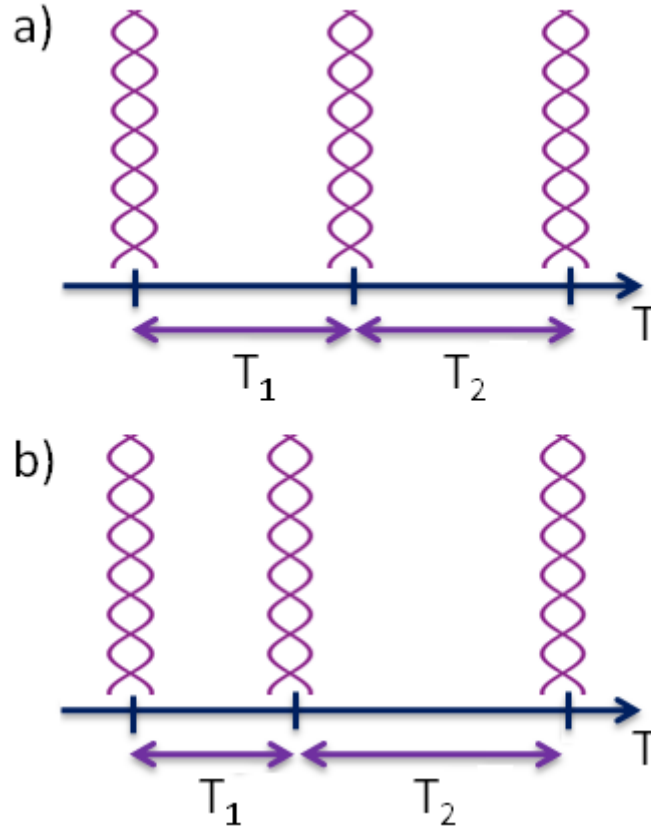


Figure 4.6.2.1: a) in the *symmetric mode*, with $T_1 = T_2$, we expect to see quantum fringe contrast, b) In the asymmetric mode $T_1 - \text{Det} = T_2 + \text{Det}$ the second laser is shifted by $\text{Det} = 200$ ns and no contrast is expected.

Depending on the experiment profile the interleaving is hardware-implemented with a duty cycle of 1:1 or 2:2 (two shots symmetric followed by two shots asymmetric), or software-coded for 1500:1500. An experiment is usually structured into a series of measurements, in-between which the experimental configuration can be changed

automatically. This way multiple interference measurements can be done in one experimental run, for example with different Talbot times, or different detuning times in the asymmetric mode. The data of each interference measurement are stored individually. The measurements are split into *sweeps*. During each *sweep* a timing configuration is sent to the pulse generators and the data acquisition is triggered in the digitizer card. In the “software interleaving mode” the laser grating timings are switched for each *sweep* (odd – symmetric, even – asymmetric). In the “hardware interleaving mode” all *sweeps* are identical. The data acquisition is done in small batches (typically 1 second long), in order to allow for a frequent laser timing correction.

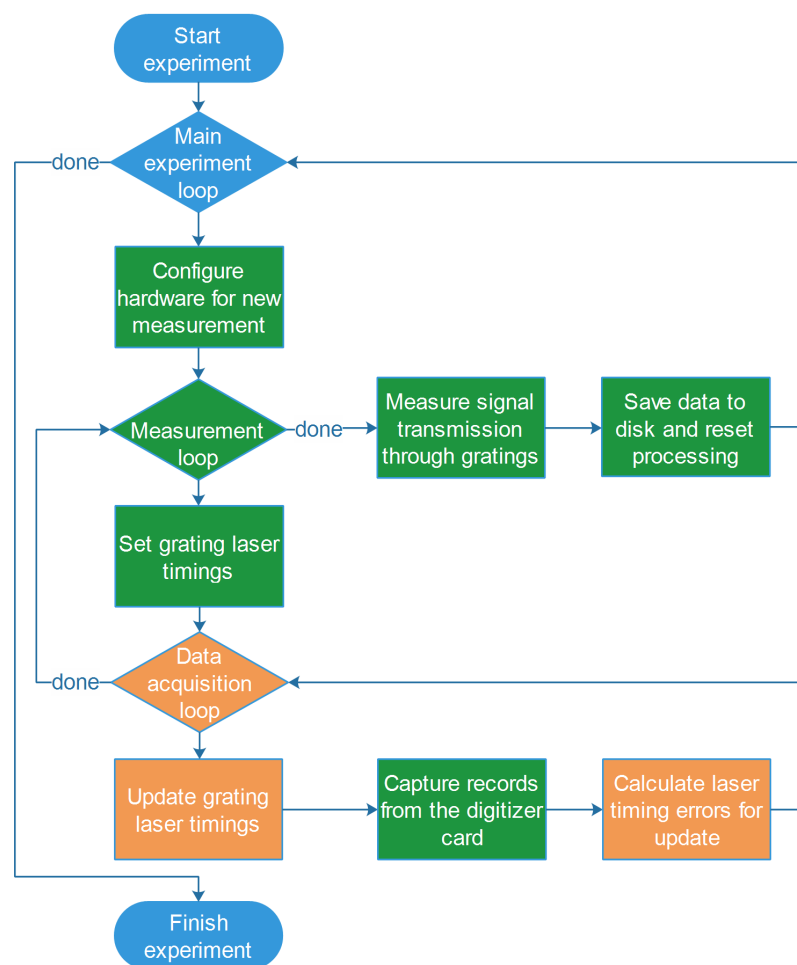


Figure 4.6.2.2: Simplified depiction of the structure of an OTIMA experiment run with MOPS. The main loop executes different interference measurements. A measurement (green) starts by preparing the experimental parameters and executing a loop which captures data for different timing modes (symmetric/asymmetric). In order to compensate for laser timing drifts (yellow) this loop in turn contains a loop which records short batches of data, monitors the timing and applies updated timings once per second. When a measurement is finished the signal transmission through each of the three laser gratings is measured. Then all captured data are saved to disk, and the data processing is reset for the next measurement.

Each record captured by the digitizer card contains multiple data. It contains the signal recorded from three photodiodes that monitor the grating lasers, the TOF mass spectrum and optionally a short sequence modulated into the signal by the pulse generators. When the mode interleaving is done in hardware there is no information in the records indicating what pulse timing configuration was active. The pulse generator therefore modulates a pulse signature into each digitizer record to indicate whether it belongs to the symmetric or the asymmetric mode.

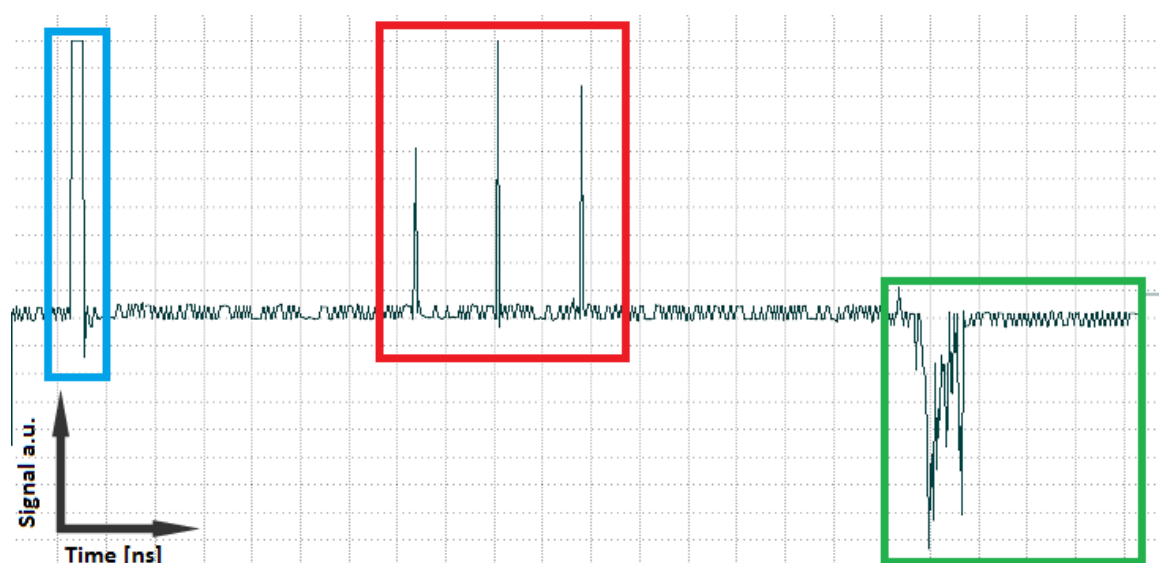


Figure 4.6.2.3: Sample of a full record containing multiple signals. The blue rectangle shows the synchronization mark, the red rectangle contains the signal from the three photo diodes, representing the laser pulses. The green one contains the signal from the TOF-MS with the mass spectrum. For example for an interference experiment with a 10-fold anthracene cluster the required data record length is about 400 μ s.

All these signals are mixed onto one channel of the digitizer card using HF Switches²⁵. This is done to save the card's onboard memory since the signals are temporarily separated anyways.

The records acquired by the digitizer card, using the "ADC" plugin, are processed in real time by MOPS. When the mode interlacing is done in hardware, first the signature must be decoded by the "FlagDecode" plugin and, as the name suggests, written into the flag list of the record. If the interlacing is done in software this step is skipped and the flag list already contains the required information. Next, the record is inspected by the "WatchDog" plugin which identifies the photo diode signals, of the three grating lasers. It compares the observed timing with the preset ideal timing, the results of these

²⁵ Mini Circuits Fast RF Switch ZYSWA-2-50DR

comparisons are saved into the flag list. There is a distinct flag that indicates whether the record should be dropped if the laser timing or power deviation is too large. The record is then passed back to the "ADC" plugin which evaluates the drop flag and, if records were dropped from the current batch, it repeats the acquisition of the missing records. The records that fulfill the conditions enforced by the "WatchDog" plugin are copied, the section with the photodiode pulses is truncated, and they are queued for processing in a separate *sequence*.

The call to the record acquisition function of the "ADC" plugin exits once the desired amount of records has been captured, or when all records were dropped. If this is the case the batch acquisition should be repeated. This acts as failsafe such that the execution does not stall if a very bad timing configuration is set. Afterwards, the script retrieves the average timing error for each laser from the "WatchDog" plugin and calculates a timing update to be applied to the pulse generators before the next batch is acquired. This way long term drifts of the laser trigger to shot delay are compensated. If it is additionally required to stabilize the laser pulse power, this can be done as well.

Once all *sweeps* are executed the typical experimental profile performs an additional measurement in which the transmission through each of the three laser gratings individually is recorded. Here, the timing of one laser is set correctly and the two other lasers are detuned so far that they don't hit the particle pulse. This is done currently without any interlacing, so there is a fourth measurement taken in which all three lasers are fully detuned.

When an interference measurement is finished all recorded data are saved to disk for further evaluation. If the experiment is set up with more than one measurement, MOPS's data processing is reset and a new interference measurement is started with a new set of experimental configurations.

4.6.2.1. Online laser monitoring and feedback

The "WatchDog" plugin is responsible for monitoring the three laser pulses which make up the interferometer and for post-selecting acquired records. The plugin searches for the photodiode peaks inside the record and extracts the time and height of each pulse. The triggering can be set such that the pulse time will be the point where the rising pulse edge starts, the middle of the rising edge is reached or where the pulse has its maximum. The trigger code is designed to ignore high frequency electrical noise that is generated by the excimer lasers when they shoot. For this reason a set of pulse selection parameters is available. The plugin implementation is generalized in such a way that it can handle an arbitrary number of lasers, even though we use in OTIMA always three. When not all expected laser pulses could be found, and also when more than three pulses have been detected, the record is marked for dropping immediately, before the timing and power are evaluated.

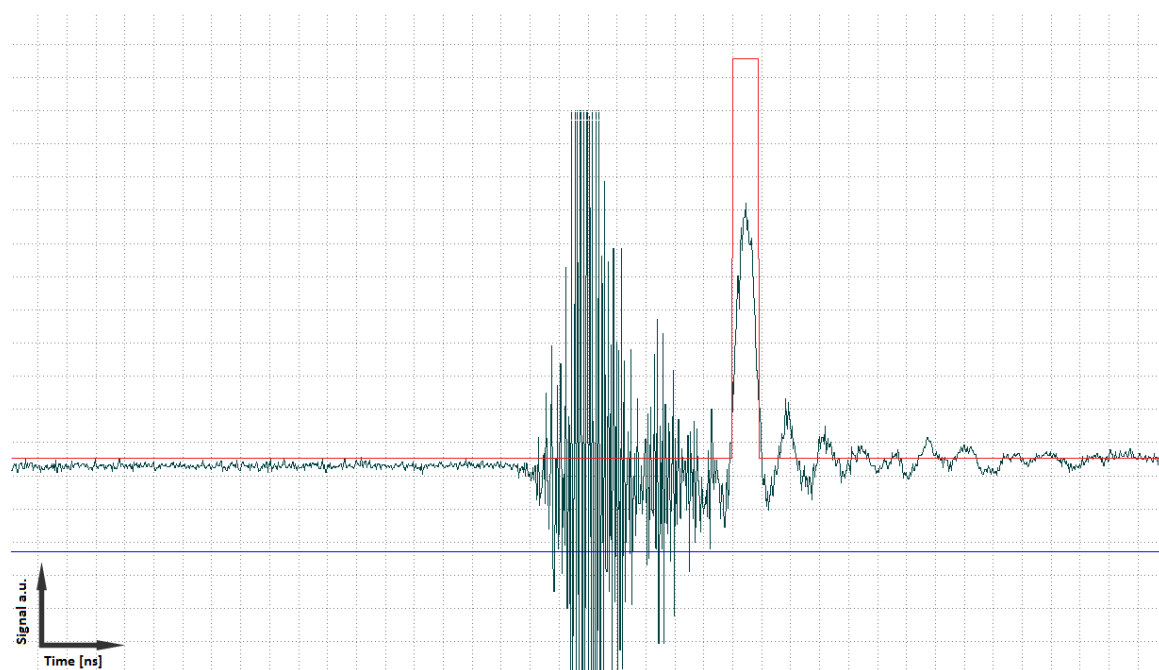


Figure 4.6.2.1.1: Example of the photo diode peak detection done by the WatchDog plugin. The dark green line shows the digitizer signal with a lot of electrical noise in front of the photodiode pulse as well as some ringing after the pulse. The plugin reliably detects the correct peak and marks it with the red line.

GAM excimer lasers are in principle capable of internal power stabilization. However, when operated with fluorine at 157 nm this feature is not available. To solve this issue the laser control software has been modified such that it can receive current measurements from the "WatchDog" plugin over the network. With these values it can stabilize the output power.

The plugin calculates two error values, one for the timing called "Jitter" and one for the laser energy called "Power". These are saved to the configuration flag list of the current record.

For the laser power a group of power classes can be preset, for example $\pm 1\%$, $+3\%$, -3% , $+5\%$, -5% . The laser power deviation as measured by the photodiodes in relation to the ideal preset value is sorted into these classes. The largest value is taken as the power error.

For the timing error value a more complex scheme is used, and there are two modes. In the default mode the plugin compares the ideal delay between two lasers with the measured delay, and saves the delay errors to a list. Then it selects the largest and the smallest delay error and subtracts them from each other. The resulting value characterizes the worst case asymmetry between the laser pulses while discarding any symmetric or common mode deviations.

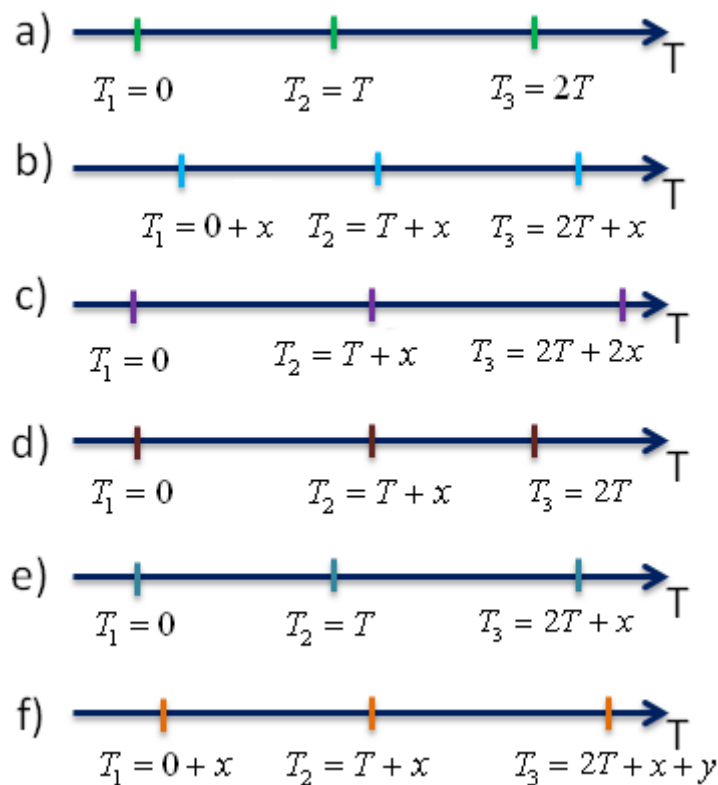


Figure 4.6.2.1.3: various examples of asymmetries, a) is ideal, b) is shifted, but perfectly symmetric. Hence, the error here is also 0, c) effectively has a different T but is still symmetric, so the measured error is still 0, in d) the error is $2x$, in e) the error is x and in f) the error is y

In the OTIMA interferometer scheme the setup is much more sensitive to asymmetries of the grating laser pulses than to a slightly different Talbot time. Common mode jitter that adds an identical shift to all three laser pulses does not influence the expected visibility. In practice, however, such a shift results in another spot of the interferometer mirror being used. If the mirror surface roughness is large enough this can cause a phase shift of the interference pattern, as described in **chapter 4.4**. The worst case being here that the used spot is so bad that the quality of one or more of the standing light waves will not be sufficient for interference.

Therefore, the plugin can also calculate the error value using a different absolute scheme. Here the error is simply set to the largest timing deviation observed in the record. This way much stronger restriction on the timing accuracy can be imposed allowing to work around issues with the used mirror. When the timing error is too large the acquired record is being marked for dropping.

4.6.3.Data pre-processing

Data records acquired by MOPS are preprocessed in real time. This allows implementing live feedback for the experiment and also helps to reduce the amount of data that needs to be saved to disk. This is done in multiple processor threads organized in plugin sequences. Using a large pool of data processing plugins it is possible to configure MOPS for different experimental scenarios.

When doing interference experiments in OTIMA the records acquired from the digitizer card are post-selected and flagged by the "WatchDog" plugin. See the **chapter 4.6.2.1** for more details. The records are truncated such that only the section containing TOF mass spectra remains. When required, additional pre-processing on the raw spectra can be done, for example using the "PeakFinder" plugin to discard electronic noise, see **chapter 4.6.3.1**.

Then the individual mass spectra are copied using the "Copy" plugin. We will come back to the copied stream later.

The next pre-processing step is to average the recorded data. This must be done in such a way that records that belong to different modes or have been flagged to be kept separate will not be averaged together. The record streams need to be split accordingly. This is all done by the "Average" plugin. It can be configured such that for each processed stream it will internally keep multiple separate *averager* instances and direct the individual records to their respective bins depending on the flag values specified in the argument line. The "Average" plugin is configured in such a way that it sums the signal rather than averaging it. Simply put, it does not do the division through the record count, but only sums the records together. The summed record count is stored as a new flag into the flag list. Thus, if a proper average is desired the omitted division can be done during post-processing. When the end of a stream is reached the "Average" plugin will return a new record stream, each record being an average of all records with matching configuration flags.

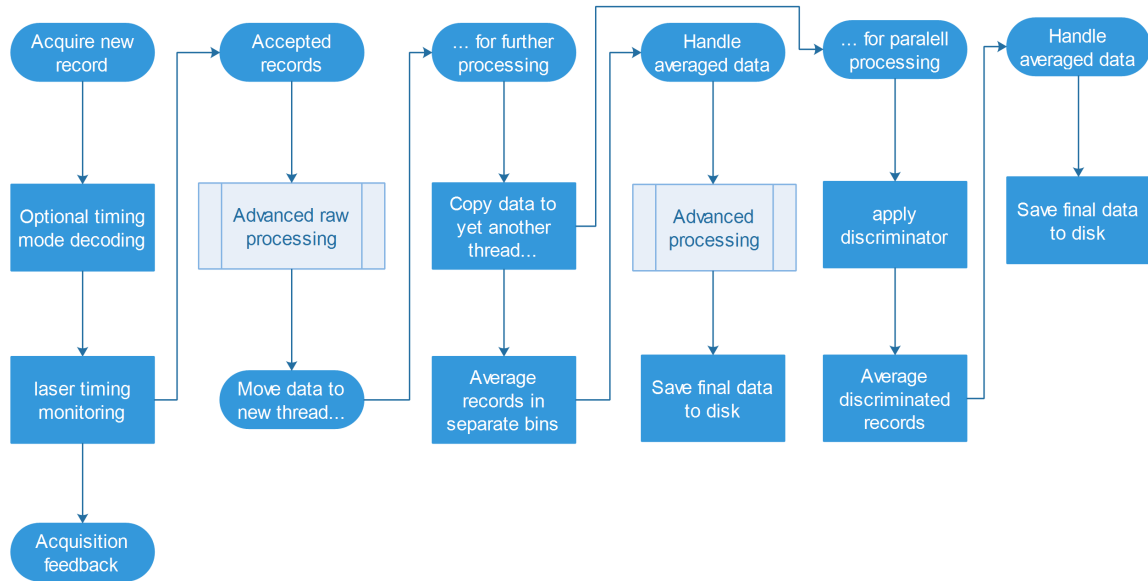


Figure 4.6.3.2: A schematic depiction of the record streaming for a simple OTIMA measurement. Each column represents an own processing thread. Data flow to right represents plugins creating new data streams. The sub processes stand for an arbitrary amount of further processing plugin entries. The records are obtained using the ADC plugin from the first digitizer channel. Afterwards they are decoded, filtered, and truncated. The ready records are averaged using the Average plugin into multiple bins based on the measured laser timings and power. Finally, the averaged data are saved to disk.

The accumulated data can be pre-processed further after this, for example, when the particles tend to get multiply charged by the detection laser, as mentioned in the **chapter 4.5**. The "MassUnfolder" plugin can be used to unfold the mass distribution, see **chapter 4.6.3.2**.

The previously copied stream is used for our error estimations, as described in **chapter 5**. The records are run through a discriminator which reduces the signal to 1 if an ion peak was present and 0 otherwise. Afterwards it is summed in multiple bins, the same way as the original signal is.

When all pre-processing steps are completed the data are saved to disk as text files using the "File" plugin. Each file contains a header section with the configuration flag list followed by the recorded data samples. During a typical interference measurement MOPS saves multiple files, containing all encountered and permitted permutations of: operation mode, laser timing jitter, and pulse power class. In addition to these information's one file containing a signal reference (no grating laser), as well as three transmission measurements where each time only one of the grating lasers hits the particle beam, is saved.

4.6.3.1. Advanced ion peak detection

There is a considerable background of electrical noise in the mass spectrum signal recorded by the digitizer card. Some of the noise originates from the digitizers ADC's itself and some of it couples in from the lab, mostly from the HV power supplies of the TOF-MS. Hence, much of the noise is made up of high frequency pulses that appear with a frequency of 20 to 200 kHz as it is typically used by switching mode power supplies. Such noise peaks have a higher amplitude than typical ion peaks, but they have a characteristic shape. Luckily, ion peaks have a different shape allowing the plugin to differentiate between them.

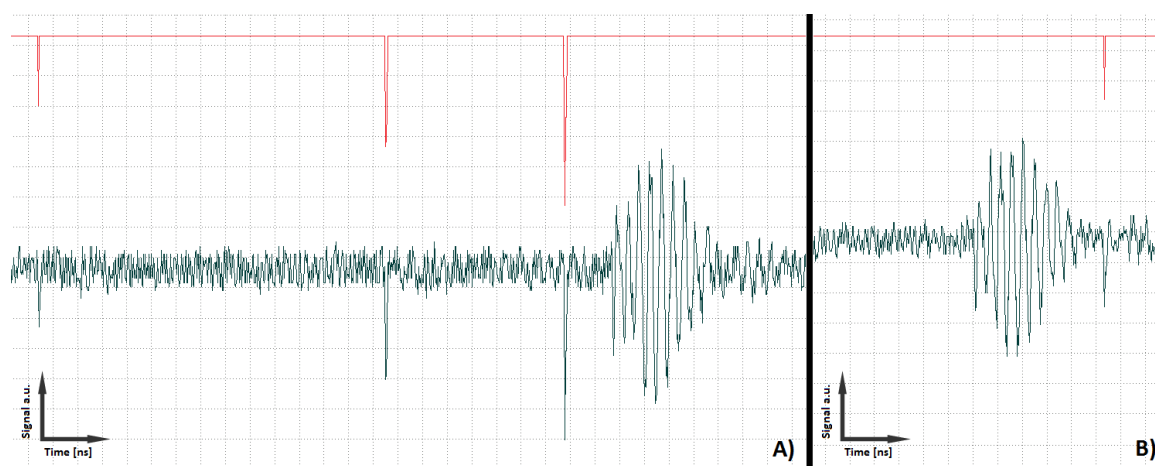


Figure 4.6.3.1.1: The dark green line is the raw digitizer signal as recorded from the TOF-MS and the red line is the processed signal. In A) there are three ion peaks and a large blob of electrical noise. In B) there is a small ion peak in close proximity to a noise blob. The “PeakFinder” plugin detects the proper ion peaks, even under such difficult conditions, and copies them into a new record. All noise outside the peaks is discarded.

The baseline noise from the ADC's is usually constant at a level in the last digit of the set digitizer range. Electrical noise that couples in has a much higher amplitude and it has a symmetric shape and is ringing at a high frequency of around 50MHz. It appears periodically and is not permanently present, in contrast to the ADC fluctuations. A typical ion peak is very different: it is a sharp pulse of 5 to 10 ns duration and peaks only in one direction. For positive ions, as used here, it goes into the negative direction due to the capacitive coupling inside the TOF-MS. The ion peak height can vary from ion to ion and also multiple ions can impact the detector simultaneously. Hence, the pulse height itself is not a distinguishing characteristic. When a lot of ions are detected at the same time the created pulse can also have some ringing. However, this is not symmetric and it decays quite fast.

The "PeakFinder" plugin uses this characteristic shapes to distinguish between different types of noise and ion peaks in real time. The plugin operates stateless, that is, it evaluates each record individually. It calculates a running average of the signal using a window size of 4k samples, for an in place baseline subtraction. While going through the records the plugin searches for a pulse above/below the baseline. If a pulse is detected it counts it and records the current sample index. Then, it looks for adjacent pulses and counts them as well. Once it counted all the pulses in the inspected sample range, it checks if the signal is symmetric or if it is strongly biased in one direction and resembles an ion pulse. If the signal passed the tests then the highest pulse in the inspected range is copied to an output record. Afterwards, the plugin continues to look for the next ion peak, until the end of the record is reached.

The main challenge while developing the plugin was to implement the pattern recognition fast enough such that the plugin could handle 250 records per second, each up to a mega sample long, in real time. The pattern recognition would not work on an averaged signal. And recording the raw data would result in 250 MB/s being written to disk, which would still need to be analyzed.

4.6.3.2. Splitting multiply charged ions spectra

The mass spectrometer used as detector in OTIMA is a TOF-MS, see **chapter 4.5**. This type of apparatus does not measure the mass m directly, but only the mass over charge ratio m/z of the ions. When the particles are ionized for the detection they can get multiply charged [130] if they have a high enough absorption cross section [131]. If a source is used that creates clusters, this may be a great issue. The "MassUnfolder" plugin was made to deconvolve the recorded spectra.

The plugin assumes that the mass of an ion is not strictly in a quadratic relation to the flight time but can also have a small linear dependency $m = at^2 + b + ct$, and some offset. With these three parameters the plugin can pre-compute the position of all singly, doubly and triply charged ions. The plugin solves the equation and determines the respective parameters when a user manually selects three ion peaks, configures the mass of the particles that make up the clusters and specifies what clusters have been selected.

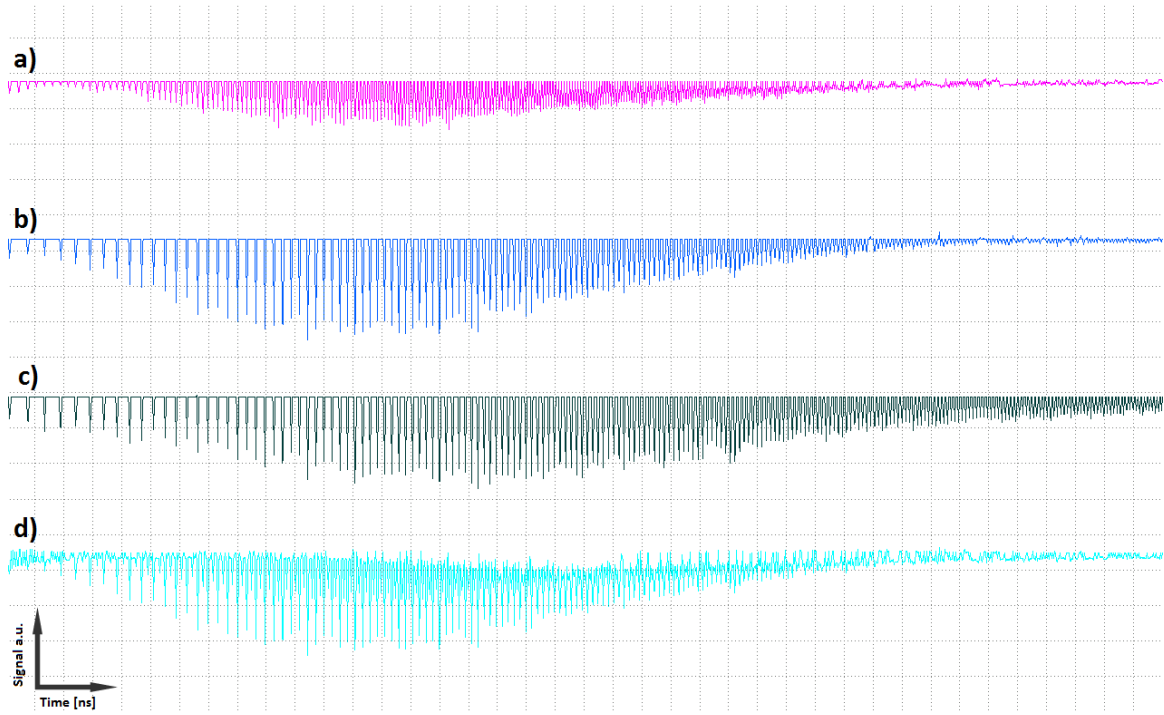


Figure 4.6.3.2.1: The cyan line (d) shows an averaged row mass spectrum captured by the digitizer card. The blue spectrum (b) contains the ion peaks at the positions of the single charged ions. The magenta spectrum (a) shows only the doubly charged ion peaks. Finally the dark green spectrum (c) shows the unfolded mass distribution.

Singly charged clusters in the TOF-MS get time slots corresponding to $t_{of} \approx \sqrt{m * n/z}$ where n is the cluster number. If the cluster is doubly charged, it gets $t_{of} \approx \sqrt{m * (n/2)/z}$. So every even peak will fall within a time slot of a singly charged cluster and every odd one will get an own time slot. With higher charges it is analogously. The plugin pre-computes these slots and splits the higher charged signal into separate spectra, each containing only the time slots used by the current charge ratio. It will approximate the values of the missing slots based on the available neighboring slots. These approximate slots will then be subtracted from the singly charged signal, such that all spectra will be separated. The higher charged spectra can be converted into singly charged ones by stretching the time axis by 2. Afterwards, they can be merged with the previously cleaned up single charge spectrum. This way no signal is lost and clean spectra can be obtained.

5. Experimental results

The data recorded by MOPS require only very little post-processing to visualize the experimental results. We load them into a MATLAB²⁶ workspace where we choose the laser timing jitter and power variation classes to evaluate. Hence, we can optimize the trade off between laser timing/power accuracy and the acquired counts even after the measurement. The selected records are averaged and we obtain two spectra, one where we expect interference contrast and one with the reference signal with detuned laser timings, see **chapter 4.6.2** for details. The MATLAB script then extracts the total signal, at a chosen mass of the relevant particles/clusters, and calculates the visibility. The total signal is the sum of all samples in a preset span around the particle mass in the TOF spectrum. The visibility is the normalized signal difference, calculated according to:

$$\Delta S_N = \frac{S_I - S_R}{S_R}$$

where S_I is the interference signal and S_R is the reference signal.

The script also sums the discriminated signal saved for an error estimation, see **chapter 4.6.3**. This gives us the amount of non-events (zero clicks in the TOF-MS) for each preset mass range, from which we extract a measure for the count rate. We assume a Poissonian distribution of the cluster counts and infer the average number of detected particles and its standard deviation ($\delta(S_I)$ and $\delta(S_R)$) from the probability of recording zero counts. Next, we use Gaussian error propagation to obtain the error of the normalized signal difference ($\delta(\Delta S_N)$).

Last but not least, the script calculates the signal transmission for a selected particle/cluster through each of the three laser gratings. The normalized signal difference is then plotted as a function of the particle mass, like in **figure 5.1**.

²⁶ MATLAB® (matrix laboratory) by MathWorks

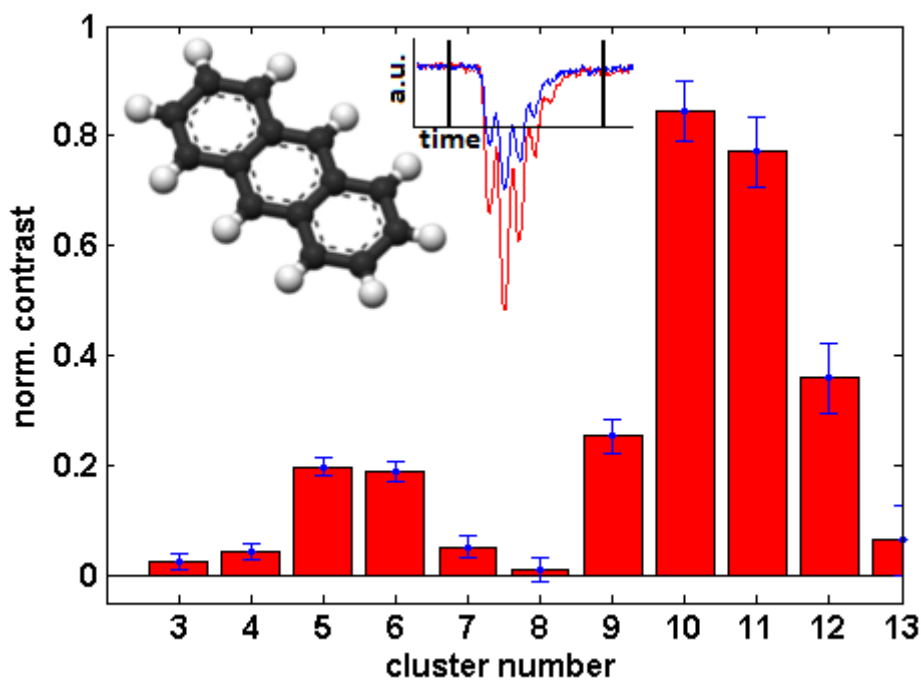


Figure 5.1: Sample of a successful interference measurement with anthracene showing 80% contrast for the 10 fold cluster. The bar graph shows the normalized signal difference for the 3rd to 13th Anthracene cluster. The included plot shows the reference signal (blue line) as well as the interference signal (red line) at the position of the 10th cluster where the most contrast is expected. A picture of an anthracene molecule is included in the upper left corner.

5.1. Ionizing gratings

We used an Even-Lavie source to create clusters of anthracene ($m=178.24$ amu), and successfully demonstrated interference in the OTIMA setup. Using argon as seed gas for the cluster creation, we get particle velocities around 690 m/s. We used a grating separation time $T = 25.9$ μ s to obtain a high contrast for the 10-fold cluster, and a detuning for the reference measurement of $\Delta T = 200$ ns. These results are shown in **figure 5.1.1**.

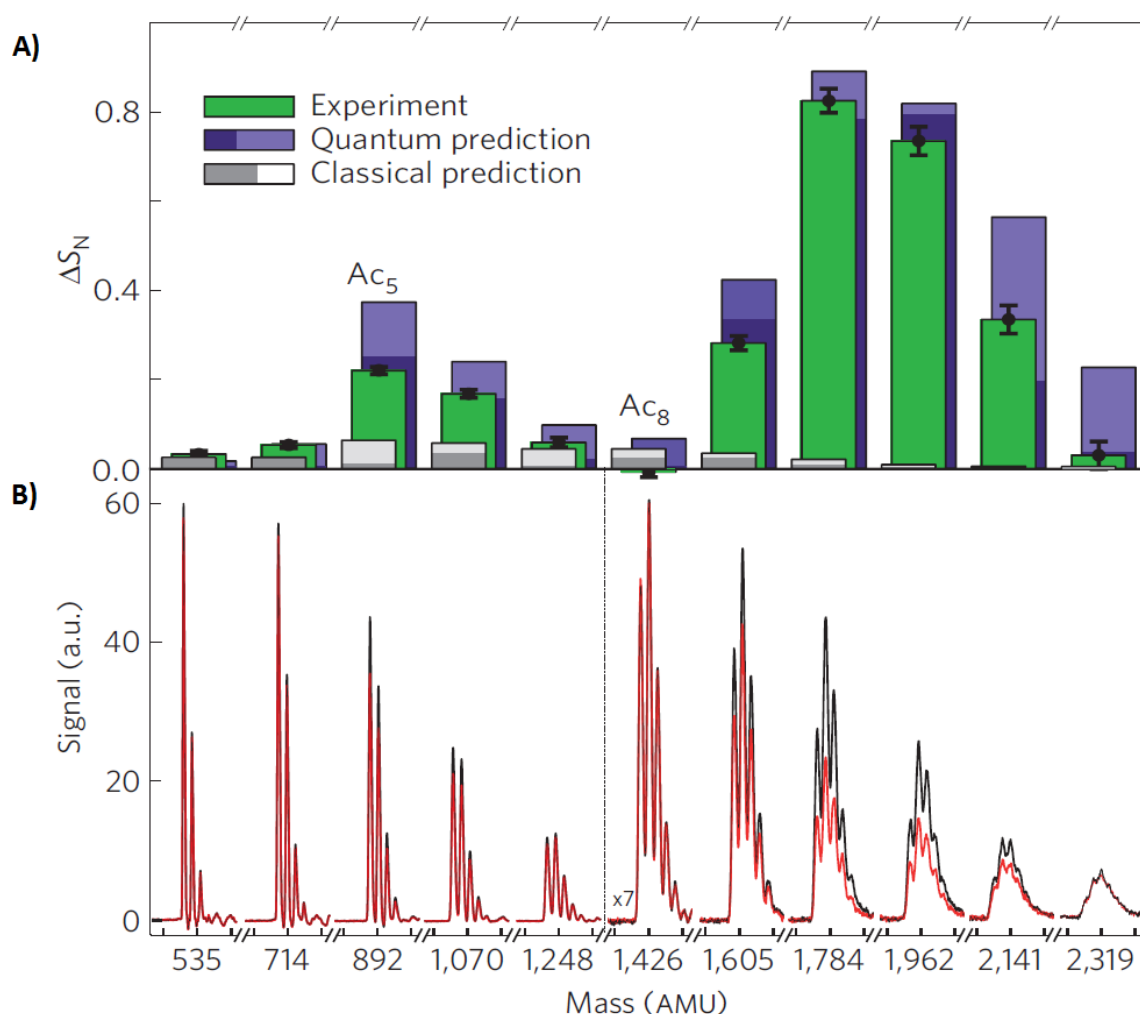


Figure 5.1.1: Anthracene cluster interference recorded in the mass spectrum (B), using argon as seed gas. The black line shows the interference measurement, while the red line shows the reference measurement. The two spectra show isotopic sub-peaks and differ for masses where interference contrast is expected. The interference contrast is represented by the green histogram bars (A). It is in good agreement with the quantum mechanical model [44] represented by the violet bars and clearly distinct from a classical model[44], depicted in gray. The light/dark distinction in the theoretical models represents a $\pm 30\%$ variation in the cluster polarizability. The figure is taken from our publication [59].

Using neon as seed gas we reach higher particle velocities of around 925 m/s and thus a shorter de Broglie wavelength λ_{db} . We used a grating separation time $T = 18.9 \mu\text{s}$ to obtain a high contrast for the 7-fold cluster. It has a mass of 1,248 amu which results in $\lambda_{db} \approx 345 \text{ fm}$. The highest mass cluster in **figure 5.1.2** has a de Broglie wave length as short as $\lambda_{db} = 275 \text{ fm}$.

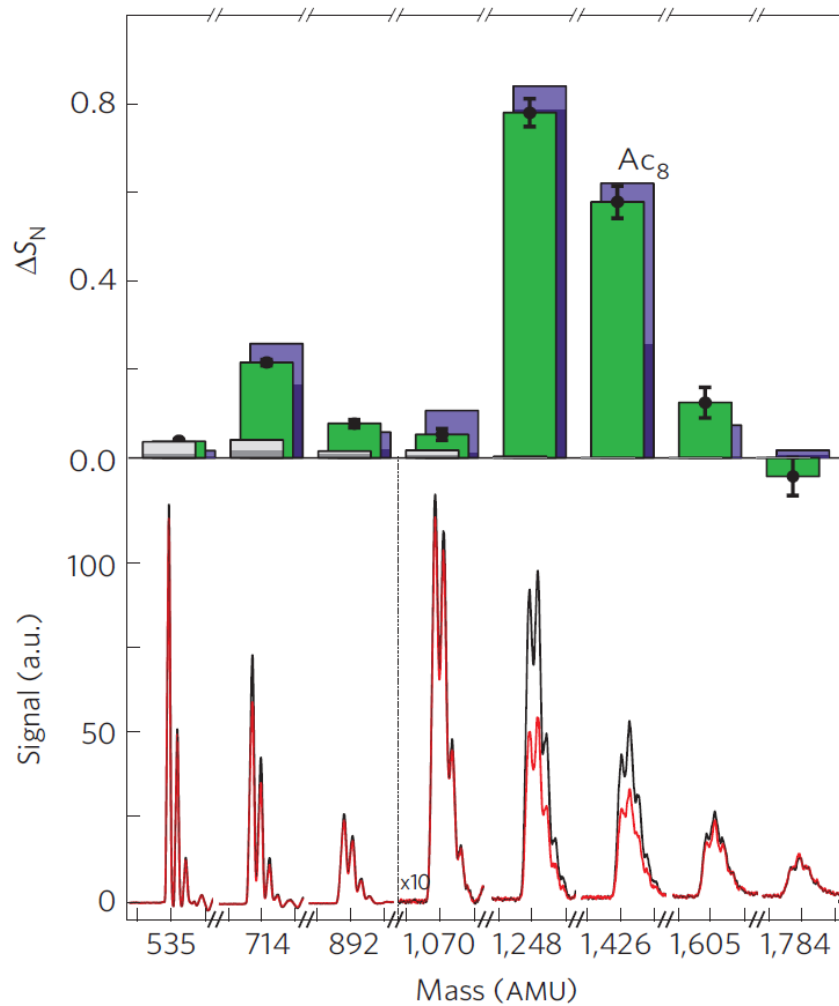


Figure 5.1.2: Anthracene cluster interference recorded in the mass spectrum, using neon as seed gas. See **figure 5.1.1** for details. The figure is taken from our publication [59].

5.1.1. Particle self-imaging acceptance time window

Particle self-imaging in a pulsed near-field interferometry setup is a process which has a very narrow acceptance time window during which the matter-waves rephase. The width is determined by the transverse momentum distribution of the particle beam [44]. In our setup it is about 48 ns long (full-width at half-maximum), as shown in **figure 5.1.1.1**. Inferring from a Gaussian fit to the data, we get a divergence angle of 2.1 mrad, which is in good agreement with the experimental setup.

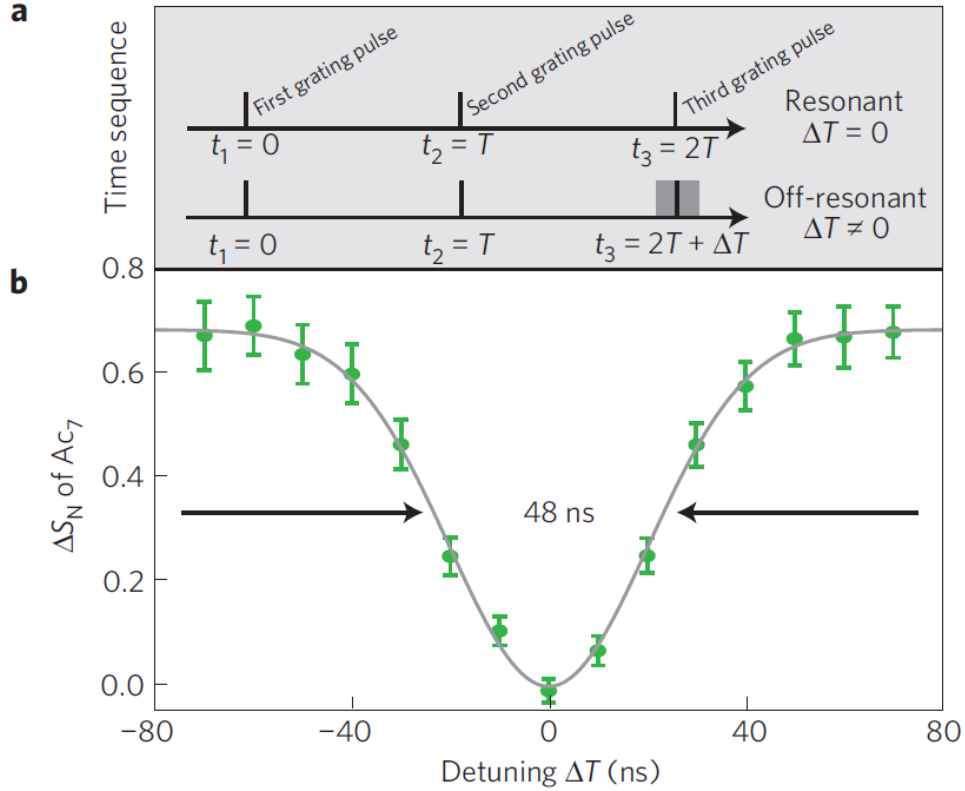


Figure 5.1.1.1: Interference visibility for the 7th Anthracene cluster as a function of the third laser grating's detuning ΔT from the ideal Talbot time T . The figure is taken from our publication [59].

5.1.2. Visualizing the interference contrast in space

The OTIMA setup allows also visualizing the interference contrast in space. This can be done by slightly tilting the second laser beam with regard to the mirror surface. As explained in **chapter 4.4**, the orientation of the standing light wave is fixed by the mirror surface. A deviation of the angle from normal incidence results only in an increase of the grating period. Here, we tilted the middle laser beam by 5.1 mrad along the particle beam, while keeping the other beams well-adjusted to about 200 μ rad. This increases the middle laser grating period by about 1.0243 pm. Changing the distance between the particle

beam and the interferometer mirror surface introduces a phase shift between the second grating and the other two. Thus, translating the position of the mirror by about 4mm allows scanning the interference pattern by one full period. Hence, we can shift the phase from constructive interference over no interference to destructive interference, as shown in **figure 5.1.2.1**. Due to the limited coherence length of the VUV lasers ($\sim 14\text{mm}$), as explained in **chapter 4.3**, it is not possible to scan much further without experiencing significant loss of the interference contrast.

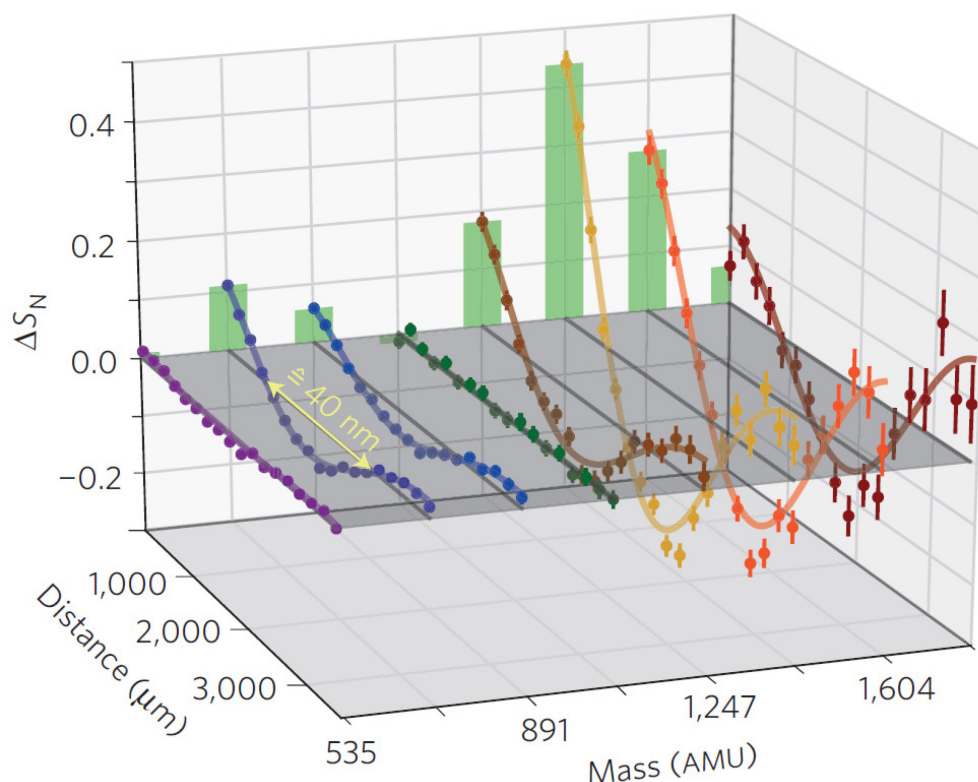


Figure 5.1.2.1: Anthracene interference visibility as a function of the phase shift (caused by changing the distance between interferometer mirror and particle beam) between the interference fringes and the third grating, recorded for the 3rd to 10th cluster. The figure is taken from our publication [59].

5.1.3. Interference with other particles

We were also able to show interference contrast for other clusters made, for example, of Caffeine (**figure 5.1.3.1**) or Ferrocene (**figure 5.1.3.2**). Thereby, we successfully demonstrated the experimental implementation of an all optical time-domain (OTIMA) interferometer and proved its versatility as a tool for quantum interferometry for a large range of nanoparticles.

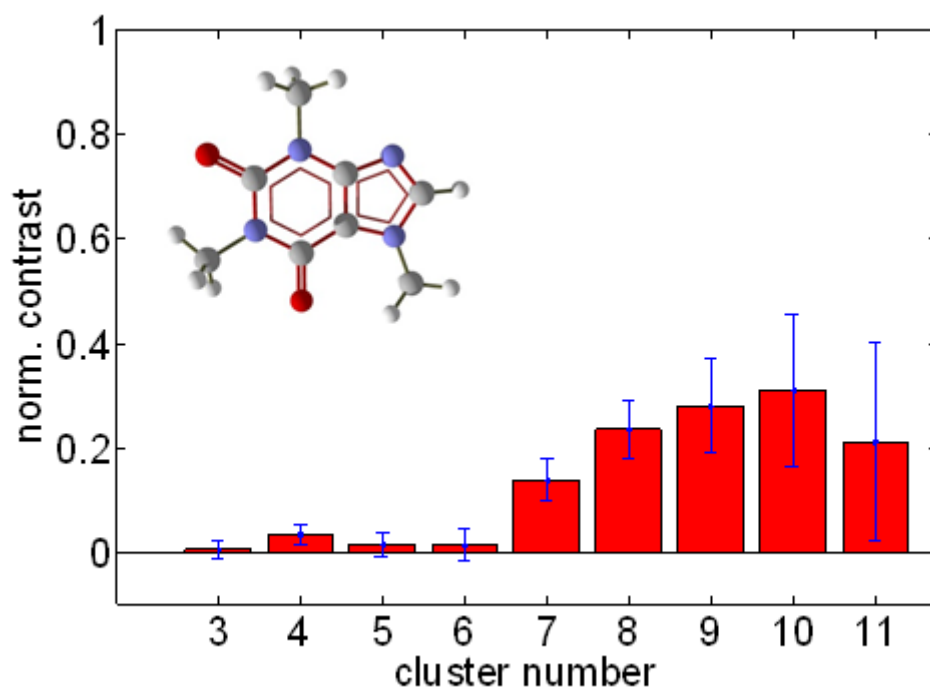


Figure 5.1.3.1: Caffeine cluster interference recorded in the mass spectrum. The bar graph shows the normalized signal difference for the 3rd to 11th molecule cluster. A picture of a Caffeine molecule is included in the upper left corner.

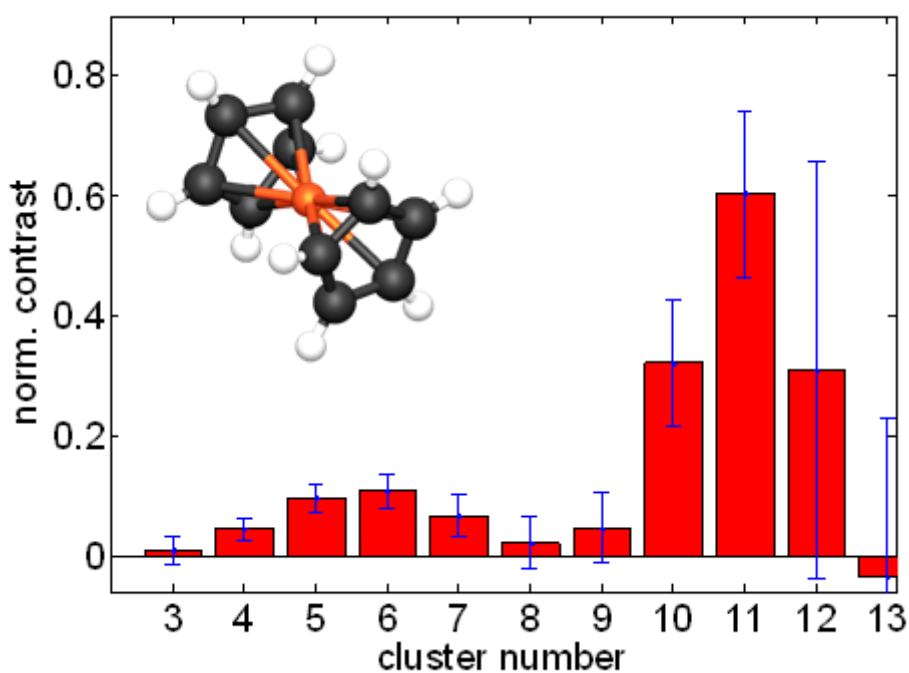


Figure 5.1.3.2: Ferrocene cluster interference recorded in the mass spectrum. The bar graph shows the normalized signal difference for the 3rd to 13th molecule cluster. A picture of a single ferrocene unit is included in the upper left corner.

5.2. Fragmentation gratings

Using the OTIMA apparatus we were able to successfully implement a new method of creating absorptive light gratings. While in our previous experiments we used single photon photo-ionization to realize absorptive gratings, this time we used single photon photo-fragmentation instead. The weakly bound van der Waals clusters as produced by the Even-Lavie source dissociate upon the absorption of a single VUV photon with an energy of 7.9 eV. Molecular dynamics simulations of such clusters done with MMFF94 [132] showed that they will dissociate within a few picoseconds after absorption. The fragments reach an escape velocity above 100 m/s which, given a forward cluster velocity of about 900 m/s, is sufficient to eject most clusters out of the 10 mrad acceptance angle of the detector. Additional later absorption events, if they occur, don't influence the grating transmission or the interference pattern.

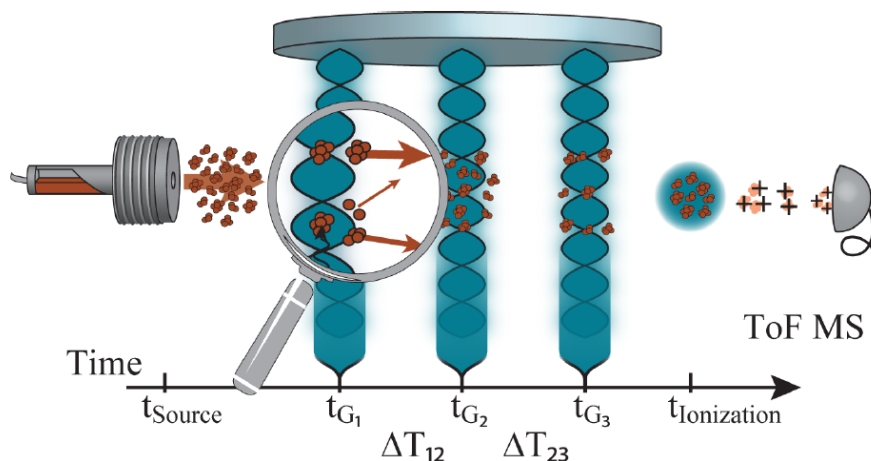


Figure 5.2.1: A schematic depiction of the OTIMA setup with photo-fragmentation laser light gratings. The clusters are produced by an Even-Lavie source, undergo photo-fragmentation in the antinodes of the standing laser beams, and the remaining clusters are ionized and detected in our TOF-MS. The figure is taken from our publication [133].

For this experiments we used hexafluorobenzene (186.05 amu) which has an ionization energy of 9.97 eV [134], and vanillin (152.14 amu) which has an ionization energy of 8.30 eV [135]. While the ionization potential usually decreases with increasing cluster size, measurements done with benzene indicated that for organic clusters it will not drop below 90% of the single molecule's ionization potential [136]. We have successfully observed mass dependent interference contrast for hexafluorobenzene clusters using a Talbot time of $T=11.5 \mu s$ tuned to the mass of the 4th cluster. Furthermore, we recorded the temporal width of the acceptance window for the rephasing of the matter-waves.

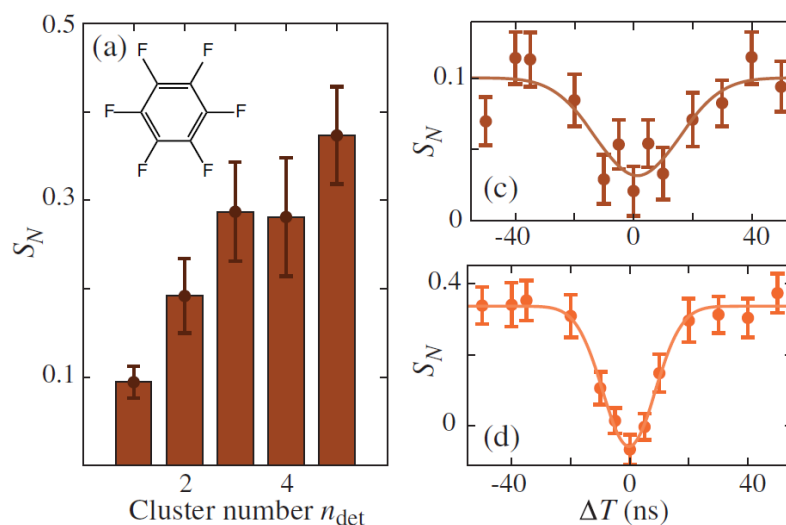


Figure 5.2.2: Hexafluorobenzene cluster interference recorded as a function of the cluster mass. And the interference visibility for the 1st and 5th cluster as a function of the detuning ΔT . The lack of clearly distinguishable Talbot orders as well as the observed interference of the monomer is due to photo-fragmentation of the clusters in the detector. The figure is taken from our publication [133].

We have also recorded interference for vanillin using a Talbot time of $T=18.9 \mu\text{s}$ which was tuned to the mass of the 8th cluster. The maxima in **figure 5.2.3** are shifted to larger clusters due to the dipole force between the particle polarizability and the laser grating light field.

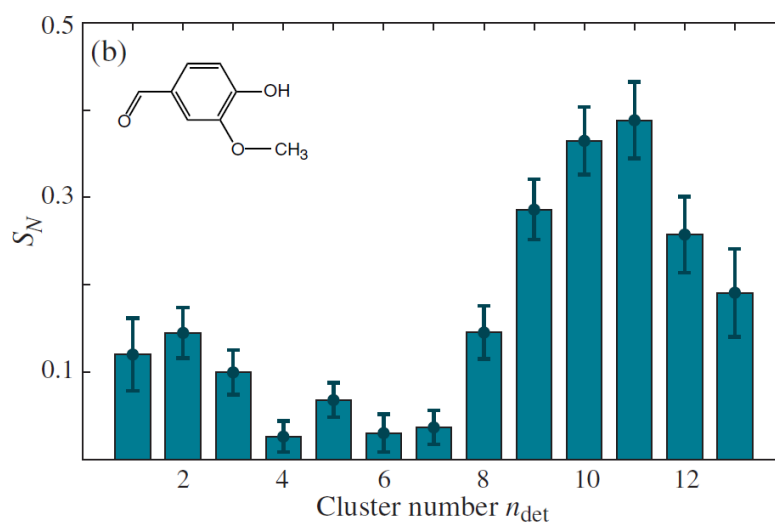


Figure 5.2.3: Vanillin cluster interference recorded as a function of the cluster mass. Interference contrast maxima appear when the grating timing is close to an integer multiple of the mass dependent Talbot time T_T of the respective clusters. The figure is taken from our publication [133].

5.2.1. Identifying the depletion mechanism

Identifying the depletion mechanism is not trivial, since ideally the detector should record only particles which haven't absorbed any photons from the gratings. Additionally, the interference pattern depends only on the grating geometry and the particles' polarizability.

In order to investigate the depletion mechanism we have recorded the relation between ionization energy in the detector and the cluster signal, as well as the relation between the signal and the depletion laser energy. For that we have moved the interferometer mirror 10mm away from the cluster beam such that, due to the limited coherence of the grating lasers, the particles will no longer experience a standing wave but only two counter-propagating running waves. With these measurements we can distinguish a single-photon effect from a multi-photon process.

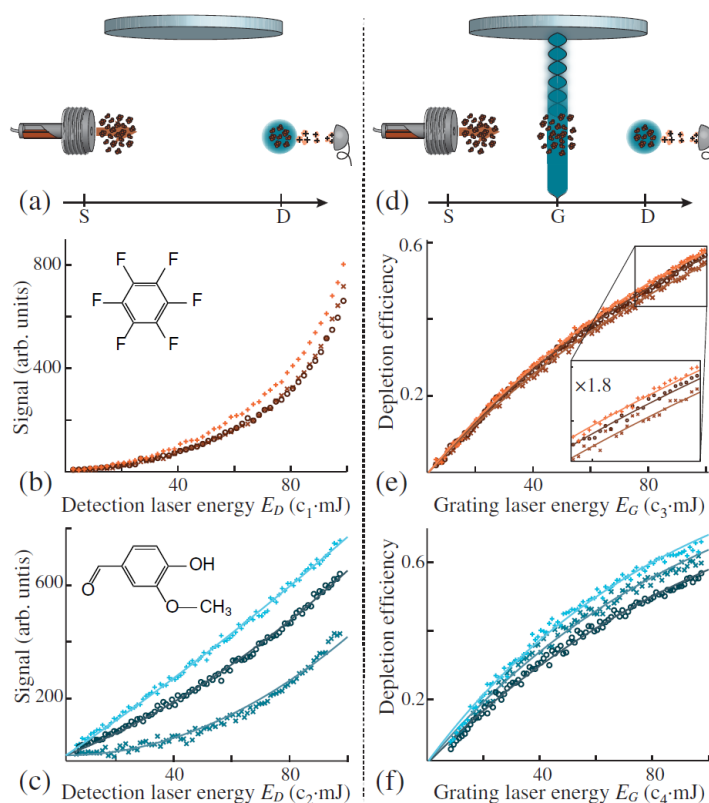


Figure 5.2.1.1: Measuring the laser power dependent characteristics of cluster ionization (a) and depletion (d). Ionization characteristics of hexafluorobenzene clusters showing a nonlinear multiphoton process for the 2nd (orange), 4th (dark orange), and 5th (light orange) clusters (b). Ionization characteristics of vanillin showing a nonlinear multiphoton process for the 3rd (blue) cluster, and a linear single photon process for the 11th (light blue) cluster as well as a transitional behavior of the 8th (dark blue) cluster (c). Beam depletion characteristics as a function of the laser power for the same clusters of hexafluorobenzene (e) and vanillin (f). The figure is taken from our publication [116].

For hexafluorobenzene we observed a strong nonlinear signal dependence on the detection/ionization laser energy, see **figure 5.2.1.1 b**. This is consistent with a resonantly enhanced photon absorption at 157 nm [137] indicating a multi-photon ionization process [138]. We have also observed a non-linear multi-photon behavior for small vanillin clusters which, however, changes to a linear single-photon one for bigger clusters, see **figure 5.2.1.1 c**. This is due to the small difference between the VUV photon energy of 7.9 eV and the molecule's ionization energy of 8.30 eV which falls with increasing cluster size, thus enabling single-photon ionization of larger clusters.

The measured beam depletion characteristics of hexafluorobenzene and vanillin, see **figure 5.2.1.1 e and f**, show exponential curves for all clusters. This behavior is expected for a single-photon depletion mechanism which has a Poissonian statistic. This demonstrates that for hexafluorobenzene the depletion mechanism in the light gratings is indeed a single-photon fragmentation process. For vanillin clusters, however, the data indicate a gradual transition from single-photon fragmentation, for small clusters, to single-photon ionization for larger clusters. The detection of intact, large clusters indicates that the photon ionization competes well with photo-fragmentation for these clusters. This stands in contrast to hexafluorobenzene where fragmentation occurs during ionization in the detector, resulting in a smearing out of interference orders and the absence of large clusters in the measured mass spectra.

5.2.2.Fragmentation gratings conclusions

With this new grating type we have extended the versatility of the OTIMA apparatus to particles with single photon ionization energies far above the photon energy in the light gratings. This enables interference experiments on very large bio-particles which have ionization energies between 8 eV and 12 eV [139] and tend to fragment rather than ionize after photon absorption [140].

6. Outlook

Our group is working towards setting a new high mass record for matter-wave interference. In order to achieve this goal the laser desorption source described in **chapter 5.1.3** is used since tailor-made particles with a molecular weight of 22,000 amu, compatible with the current OTIMA setup, were observed to fly slowly with a similar source [104]. Particles in this mass range require a Talbot time of about 330 μs in order to create a first order interference pattern. Such an experiment is not only demanding with respect to the interference itself, but also with respect to the data processing. In order to estimate the data storage capacity and the data rates needed to perform such an experiment, we start from the total record length which will be twice the Talbot time, plus the time from the interferometer to the detector: 850 μs . To that the time needed to record the mass spectrum: 200 μs , must be added. This sums up to almost 2 ms. Using again a timing resolution of 1 ns and a voltage resolution of at least 10 bit this results in data records of 4 MB length accumulated with a repetition rate of up to 250 Hz, i.e. a maximal total data rate of 1 GB/s.

The currently employed digitizer card, however, has an on-board memory of only 1 MSamples and its peak transfer rate is limited to 250 MB/s. Thus, it became necessary to upgrade the setup with a new digitizer card²⁷, which has a memory of 350 MSamples and a peak data transfer rate of 3.6 GB/s. Additionally, the new hardware has a FPGA²⁸ (Field Programmable Gate Array) on board, which allows to process data directly in hardware. Given that for these Talbot times the largest part of a data record contains only sparse data, see **chapter 4.6.2**, implementing some of the "WatchDog" plugin functionality in the FPGA appears advantageous. The data acquisition plugin has already been developed for this new digitizer hardware.

²⁷ SP Devices ADQ412 – 2/4-channels, 12-bit, 4 GS/s

²⁸ Xilinx Virtex 6 LX240T

6.1. Functional measurements on complex bio molecules

Matter-wave interferometers have been shown to be sensitive tools for measuring particle properties [105, 106, 141, 142]. This is due to the fact that the interference patterns are highly sensitive to forces acting on the particles while they are crossing the interferometer. Particle deflections even by a fraction of the grating period can easily be resolved by observing the changes in the interference patterns. Here, we must distinguish between interactions which move the patterns (phase-shifts) and those which smear out the contrast (decoherence).

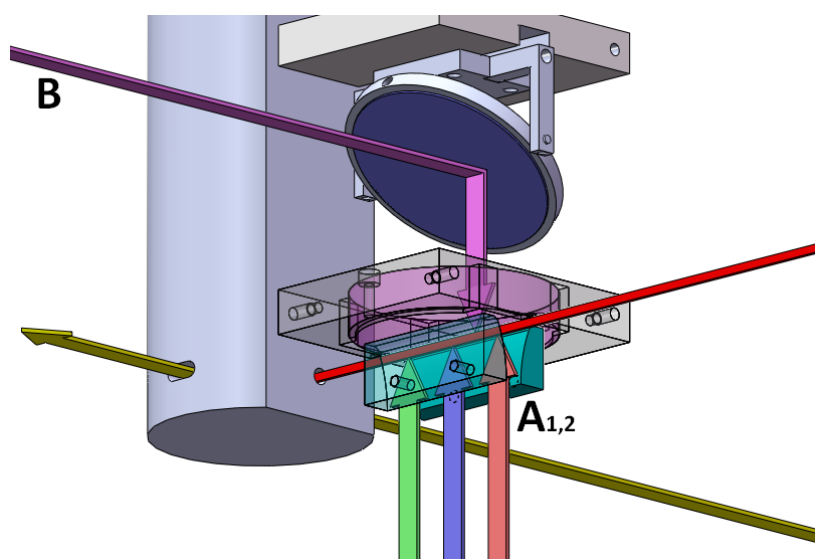


Figure 6.1.1: Interferometer assembly with two symmetric deflection electrodes added around the particle – laser interaction volume. A_{1,2}) Deflection electrodes for deflectometry, B) Recoil laser for absolute absorption spectroscopy, for other details see **figure 4.4.4**.

Matter-wave interferometry with complex biomolecules is challenging as it is generally considered difficult to bring large neutral molecules intactly into the gas phase. With the source presented in **chapter 4.1.4** we have successfully created beams of cold, neutral polypeptides. With the GBMS setup it will be possible to perform functional measurements with the OTIMA apparatus on bio-particles up to ~8,000 amu.

6.1.1. Measuring static polarizability

Static polarizabilities of particles provide information about their structures and conformations [143]. By combining a Stark deflector with a Talbot-Lau interferometer the static polarizabilities α can be measured [105, 141]. This is an assembly of two

specially shaped electrodes which provide an inhomogeneous electric field. This E field is formed such that it provides a homogeneous force $F = \alpha(E\nabla)E$ in the region where the particle beam passes the deflector. This force deflects a particle by:

$$\Delta x \approx \frac{\alpha}{m}(E\nabla)E\left(\frac{l}{v}\right)^2$$

with m being the particle mass, v its velocity and l the length of the electrode it is passing. In previous setups [105, 141] the deflection electrodes have been placed between the first and second grating. Hence, in addition to the deflection Δx there will also be some influence from stray fields before and after the electrode assembly. In an all optical interferometer it is, in principle, possible to use electrodes which cover the entire interferometer, see **figure 6.1.1**. Thus, the effect of stray fields at the electrode edges should be minimized. Also, in the time domain the velocity dependence is gone and the deflection is given by:

$$\Delta x \approx \frac{\alpha}{m}(E\nabla)ET_T^2$$

where T_T is the Talbot time. Hence, the precision of the measurement is no longer limited by the velocity selection. The static polarizability can be calculated from the recorded shift of the interference pattern as a function of the electrode potential.

6.1.2. Measuring absorption cross sections

Measurements of absorption cross sections are typically done using vapor cells [144]. This requires the analyte particles to have a sufficient vapor pressure to form a gas with a measureable opacity. In order to perform absolute measurements the vapor pressure must be accurately known. This is, however, usually not the case for complex molecules.

Using a matter-wave interferometer allows to measure the absolute absorption cross-section with high accuracy [106]. A probe laser applied between the first and second grating is absorbed by some of the particles, see **figure 6.1.1**. Hence, the particles will receive a momentum kick which changes their position with regard to those that haven't absorbed any photons. These random shifts are diminishing the contrast of the interference fringes. From the changes of the fringe visibility as a function of the probe laser intensity the absolute absorption cross sections can be extracted.

6.2. Interferometer mirror upgrade

To mitigate the mirror inhomogeneity described in **chapter 4.4**, which currently forces us to limit the detected extent of the particle beam to about 1 mm, OTIMA will be upgraded with a new interferometer mirror. It is rectangular and has a clear aperture of 50×30 mm, while the old round mirror head a clear aperture of about 45 mm in diameter. This will allow us to study slightly faster particles as the grating lasers can be further apart, see **figure 4.1.1** in **chapter 4.1**. Additionally, the rectangular shape allows using different spots of the mirror by moving it laterally. The mirror has a thickness of 20 mm in order to minimize deformations.

The new mirrors have been coated by LASEROPTIK GmbH in cooperation with Hellma GmbH who provided the polished CaF_2 substrates.

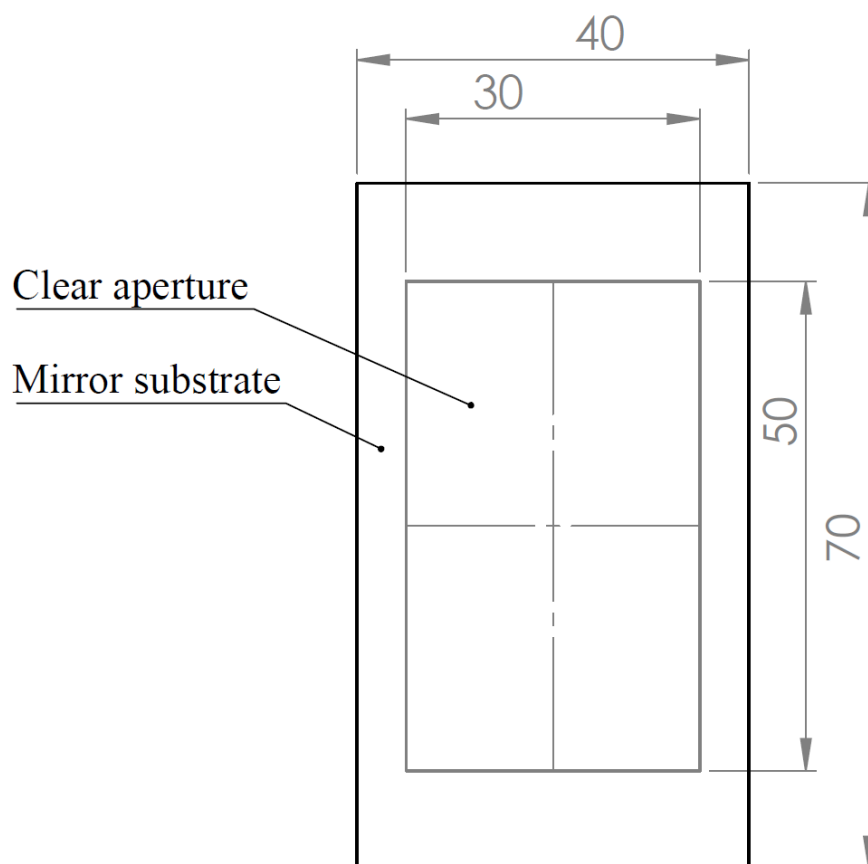


Figure 6.2.1: The new main OTIMA mirror used as a common boundary for all three standing light waves acting as optical gratings.

The new mirror has a reflectivity of 95% at normal incidence and the surface height variation from peak to valley in the clear aperture is about 12 nm. The reflectivity is slightly less than that of the current mirror, but in exchange its flatness is five times better.

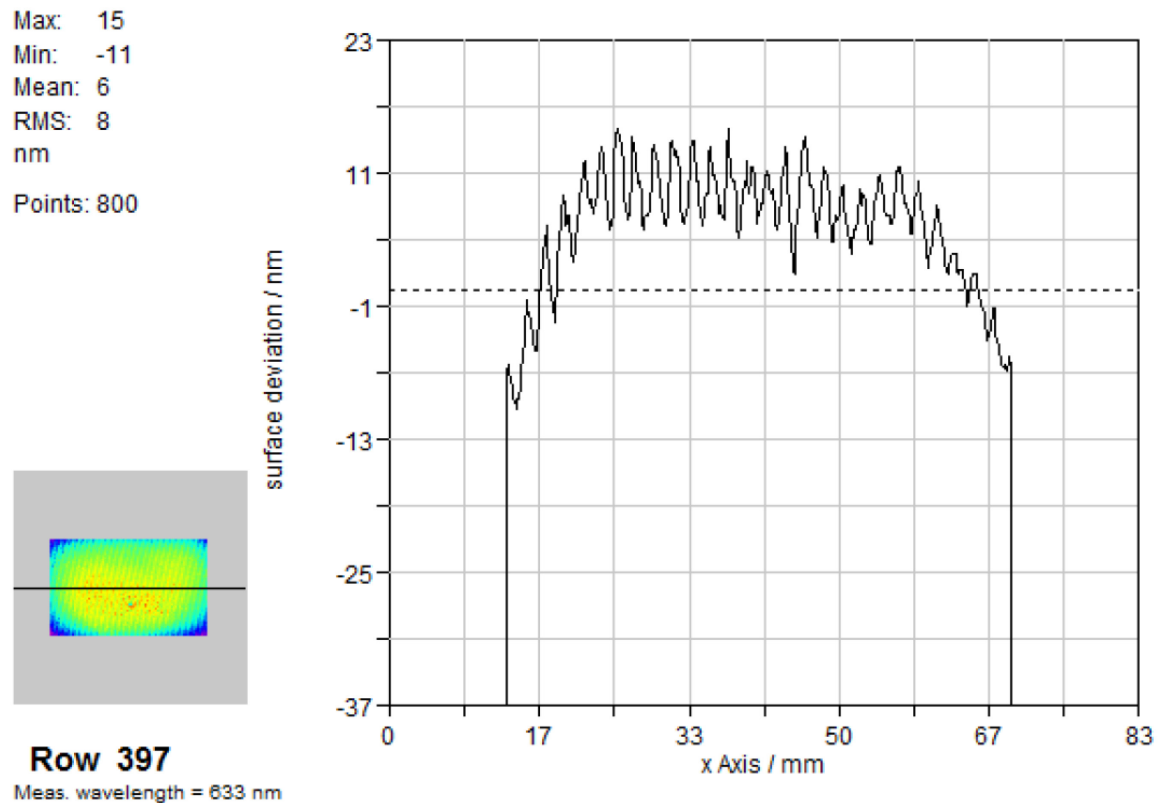


Figure 6.2.2: Interferometric measurement of a new OTIMA mirror at 633 nm done by LASEROPTIK GmbH.

With this new mirror the phase shifts introduced by the surface roughness will no longer be a major limiting factor for the interference contrast. The drawback of a slightly higher running wave contribution [56] due to the lower reflectivity, and thus a higher background, is more than compensated by the signal increase gained from the ability to use larger portions of the particle beam.

6.3. Interference with large metal clusters

The magnetron sputter source presented in **chapter 4.1.1** is capable of providing the heaviest particles of all the tested sources. With a similar setup [145] continuous flow rates of 5×10^6 clusters/second with a mass of 2×10^5 amu have been observed. The signal loss due to the required chopping of the beam as well as the low total transmissivity of the interferometer made the required duration of a measurement, prohibitively long. Large metal clusters still remain, due to their high density, good candidates for matter-wave interferometry in the 10^5 amu range and beyond.

For example, a 10^6 amu gold cluster which has a density of 19.3 kg/m^3 would have a diameter of only 5.5 nm. Thus, it would still fit well through OTIMAs optical (period $d=78.8 \text{ nm}$) gratings which, at a laser intensity required for good contrast, have a “slit width” of about 18 nm, see **figure 3.2.2.1**. Biological particles with a density of only $1.2\text{--}1.3 \text{ kg/m}^3$ would grow to already 13.6 nm for the same mass.

A possible solution could be to use an RF ion trap [146, 147] to accumulate negatively charged ions coming from the source. Once trapped the clusters could be cooled down to 10 K using buffer gas [148, 149]. The main limiting factor here would be the space charge limit of such a trap [150, 151], i.e. the amount of ions fitting into a trap of a given volume, typically about $10^6/\text{cm}^3$. Quickly extracting ions from a large trap would require accelerating them in the forwards direction, which counteracts the cooling. On the other hand, without forward motion the free particles would spread in all directions.

A new experimental setup could perform the interference measurement inside the ion trap. The first grating laser pulse would implement an inverse grating, i.e. particles which are neutralized by photo-induced electron detachment would fly on through the interferometer while the remaining charged particles would be discarded, see **chapter 3.2.2**. After a first Talbot time a second pulse would diffract the particles, like in the current OTIMA scheme. Finally, the third grating pulse would probe the interference pattern by ionizing the particles again. Afterwards the ionized particles would be trapped: that would correspond to an inverse grating. Alternatively, they would be discarded and the remaining neutrals post ionized and trapped: this would be a regular grating. Since ion traps can be made mass selective [152, 153] a mass resolving detector would no longer be necessary. Hence, the ions could be directly detected by guiding them onto a MCP.

7. Appendix

The measurement and control software used in OTIMA, MOPS (Molecular Optics Programming System) is a complex custom development. In this appendix a superficial overview of the application is presented. The principle of operation of the custom script engine will be explained as well as the debug mechanism and the function binding scheme. Afterwards, we will take a look at the high speed parallel data processing mechanism. Last but not least the plugin interface specifications will be described in full detail.

7.1. MOPS script engine

The MOPS script engine implements a simple *Turing complete* programming language. The only data type the script uses is *string*, but if needed a more sophisticated *variant type object* could be used. The script is organized into functions and all arguments are passed *by reference*. That means that if the argument is changed inside the called function, the variable passed by the caller will also be updated. Hence, any amount of parameters can be passed back to the caller. Each function has an own *heap* of local variables and can access a shared *heap* of global variables. A variable is created on the global *heap* if it starts with a point, otherwise it is either a passed argument or is created on the local *heap*. Unlike in most programming languages the arguments are not identified by order, but by name. That means instead of:

```
SomeFunction(Arg1, Arg2, Arg3, ...)
```

Code 7.1.1: Typical function calling convention, like it is used in C/C++, JS or Python. The arguments are separated with comas and their order is predetermined in the functions signature.

a call looks like this:

```
SomeFunction(Param1 = Arg1, Param3 = Arg3, Param2 = Arg2, ...)
```

Code 7.1.2: MOPS script function calling convention. The parameter order is arbitrary. The parameters are identified by names that are used inside the function.

The called function can be either a function defined inside the script or a *binded* API function that is a natively implemented function exposed by the engine. The arguments can be specified in an arbitrary order and left out if they are not mandatory for the particular function. Inside the function the passed arguments are referenced by their names as specified in the call code. The presence of a variable can be tested by prepending the variable name with a `#`. The expression will evaluate to `true` if the variable is present in the argument map or in the local or global *heap*, and to `false` otherwise. A variable can not only be accessed by a hard coded name but also by evaluating another variable.

```
VariableName = "Param" & 1  
TestValue = [VariableName]
```

Code 7.1.3: Accessing a variable by a custom name is done by putting the variable which contains the name into square brackets.

In this example the variable `TestValue` will be assigned the value of the argument `Param1`. The script also allows using a variable as function name. This emulates the usage of function pointers. When evaluating equations all standard operators are available. In addition to these operators there are built in functions available that can be used for more advanced calculations. The script exposes all standard math functions available in `<math.h>` as well as some convenient custom functions. The equations can contain brackets `( )` and are internally ordered according to the operator precedence before evaluation.

The script compiler knows only simple expressions like labels, `goto` commands, function calls and operations on variables. To use flow-control structures such as `if`, `else`, and `loop` a preprocessor was implemented that converts these into simple conditional `goto` operations, as shown in the following example:

```
Function SomeFunction Begin  
    LoopCount = 0  
    loop (.RunLoop)  
        LoopCount + 1  
        LogLine(Line = "Loop" & LoopCount)  
        if (LoopCount == LoopsToMake)  
            break  
        end  
    end  
Function End
```

Code 7.1.4: This example shows a simple MOPS script as it would be entered by a user, in this particular case it is a function that repeatedly writes some text to the log.

```

Function SomeFunction Begin

    LoopCount = 0

    loop_1:
    goto !(.RunLoop) end_loop_1

        LoopCount + 1
        LogLine(Line = "Loop" & LoopCount)

        goto !(LoopCount == LoopsToMake) end_if_2
            goto end_loop_1
        end_if_2:

    goto loop_1
end_loop_1:

eof:
Function End

```

Code 7.1.5: This is the same script as in the previous code block but after it has been run through the preprocessor. All complex flow control structures have been replaced with simple `goto` statements. The code is now ready to be compiled into byte code.

In this example `.RunLoop` is a global variable that, if set to false, will abort the loop, and `LoopsToMake` is an argument passed by the calling function that specifies how many loops to execute.

The equation execution is done using an *accumulator architecture*. This means that every operation like, addition takes one argument, and the other argument is always the accumulator variable. To illustrate this let's go through the following simple example:

```

Equation in script
x = a + b + c / d + e * f

Equation after conditioning
x = (a + b + (c / d) + (e * f))

Equation evaluation step by step
temp_acc_1 = a
temp_acc_1 + b
temp_acc_2 = c
temp_acc_2 / d
temp_acc_1 + temp_acc_2
temp_acc_3 = e
temp_acc_3 * f
temp_acc_1 + temp_acc_3
x = temp_acc_1

```

Code 7.1.6: Example of equation execution in MOPS, first the equation is sorted according to the operator precedence, then it is converted into a list of simple instructions, and then it can be executed.

The first expression in an equation must be a variable and is the accumulator. This is then of course followed by an operator and an argument expression. These can be either another variable or another equation. If it is a variable then the operation is executed, if it is another equation the equation is executed first. In that case a new accumulator is created internally and initialized with the first variable. Once the sub-equation is evaluated the sub accumulator is used as argument for the parent equation. So in our example before anything is done to `x`, the whole bracket is evaluated. One could of course use `x` as accumulator directly, not putting the entire equation into brackets, and save one temporary accumulator. In this case, however, the following example would fail:

```
Equation in script
x = 10
x = 20 + x

Equation evaluation step by step
x = 10

x = 20
x + x
```

Code 7.1.7: Example of a faulty equation conditioning. Here, the content of the `x` variable is overwritten before it is used, and the result of the operation is not 30 as one should expect but 40.

The accumulator design becomes apparent for example when incrementing a variable by one, like in the preprocessor example for `LoopCount` the user just writes `LoopCount + 1`. Here `LoopCount` is the accumulator on which a sum operation is executed with a constant argument 1. When calling a function with an equation as argument, a temporary accumulator is passed to the function. This mechanism can be used to pass arguments to the function *by value*. That means in a way that if the function changes the argument this change will not affect the variables of the calling function. Like in the following example done with `Param2`:

```
SomeFunction(Param1 = Arg1, Param2 = (Arg2), Param3 = 123,...)
```

Code 7.1.8: MOPS function calling convention with a constant argument. Placing the argument in brackets makes it an equation for the script and a temporary copy of `Arg2` is passed to `SomeFunction` instead of the actual `Arg2`. Also any constant arguments are passed as temporary copies, like `Param3`.

The virtual machine is implemented in such a way that the script execution can be suspended and the control returned to the calling thread, while all internal machine states are maintained. This way the thread can resume the script execution where it was interrupted. With this mechanism a *synchronously* written script can call *bounded* API functions which execute *asynchronously* and once the function finishes the script is resumed. This is advantageous for the users as *asynchronous* programming is generally considered much more challenging, and with this mechanism it can be avoided from the user perspective. It is also used by the debugger mechanism. If the debugger is attached, a *preemption* timer interrupts the script execution every 100ms and returns the control to the debugger. It then can inspect the *call stack*, access local and global variables and set breakpoints. If the script runs into a breakpoint the control is immediately returned to the debugger which can execute the typical commands like: execute single instruction, skip instruction, enter function, execute until function return. When the execution is suspended the debugger can also execute single script lines entered by the user in the context of the selected function. This is a very convenient debugging and development tool as it allows the user to control the experiment by hand while it is running.

The script uses a simple *function binding* scheme, to add a API function `RegisterFunction(const wstring& FxName, FX Fx)` is used. `FxName` is here the name of the function from which it will be called from script, and `Fx` is a pointer on the static C++ function that is to be *binded* to the engine. The function must have the signature `int (*FX) (VarPtrMap* pArgMap, SContext* pContext)`. The `int` return value is an error code. If it is not 0 the execution will be interrupted and the control returned to the calling thread. Return codes > 0 indicate a resumable state like an interrupt, a preemption break or a breakpoint, and values < 0 indicate an error. The `pArgMap` argument is a map of the function call arguments as issued by the script. `pContext` is a structure containing engine information and providing access to a context object that can be passed when starting the script execution. With this a static API function can operate on objects of which there is more than one instance.

7.2. MOPS record streaming

The processor script is made up of process blocks, each one being executed in an own thread and representing one processing sequence. A block starts with a `PROCESS BEGIN` statement where a record is pulled from a plugin. Then there is a sequence of plugins that process the record stream. And finally there is a `PROCESS END` statement where optionally the record can be pushed to another plugin. If the end statement is empty the processor deletes the record. Each record pull, process, or push operation starts with the plugin name, is followed by an optional stream ID separated with a dot from the plugin name and can be followed by a string containing parameters for the plugin configuration. The argument line can contain `%Text%` statements. Here the text between the `%` signs is a name of a configuration flag and it will be replaced with the actual value of the flag from the current record, before being passed to the plugin.

```
PROCESS ADC.CH1 BEGIN
    Copy.stream1
PROCESS Copy.stream2 END

PROCESS Copy.stream1 BEGIN
    ' do something with the copy of the stream
PROCESS END

PROCESS Copy.stream2 FILTER:FLAG=Value1 BEGIN
    ' handle the records where the flag with the name FLAG is set to Value1
PROCESS END

PROCESS Copy.stream2 FILTER:FLAG=Value2 BEGIN
    ' handle the records where the flag with the name FLAG is set to Value3
PROCESS END
```

Code 7.2.1: Simple example of copy plugin usage, here data are obtained by the digitizer card from channel 1, copied, and queued as `stream1` and the original data are queued as `stream2`. In parallel `stream1` is pulled and discarded, while stream `stream2` is filtered and pulled into two separate threads.

Plugins can maintain internal states and process the records in the context of these states. A state may be linked to a single stream or a set of streams. Alternatively, they can also operate without maintaining state information for the stream, i.e. handling each record individually.

```

#define SAMPLE_BYTE 0
#define SAMPLE_INT8 1
#define SAMPLE_INT16 2
#define SAMPLE_INT32 3
#define SAMPLE_INT64 4
#define SAMPLE_DOUBLE 5

struct SSegment
{
    byte      SampleType;
    void*     ptrArray;
    DWORD     arrSize;
    SFlag*    FlagList;
    int       FlagCount;
};

```

Code 7.2.2: Each record handled by the plugins is implemented as a structure `SSegment` containing a byte array with the actual data, a size field indicating in bytes how much data there is as well as a type field specifying the used data type. The structure also contains a list of configuration flags of the type `SFlag`.

```

struct SFlag
{
    wchar_t   Name[16];
    SArgument Flag;
};

```

Code 7.2.3: A flag structure contains a name, which can be up to 15 characters long, represented as a 0 terminated wide string and a `SArgument` object containing the flag value.

Most plugins were designed to operate with data as recorded by a digitizer card. These are usually arrays of 8 or 16 bit long signed integers. However, also longer data types, such as a double precision floating point type, have been implemented and can be handled by most plugins. All these standard types can be natively previewed in the measurement view. There is also a binary data type that can be used for custom data.

```

#define ARG_BYTE 0
#define ARG_INTEGER 1
#define ARG_REAL 2

struct SArgument
{
    char Type;
    union uArgument
    {
        byte Bytes[64];
        int64 Integer;
        double Real;
    }
    Argument;
};

```

Code 7.2.4: The argument structure contains a type field indicating its data type and a *union* that can be accessed either as a 64 bit long integer a double precision floating-point number or a 64 byte long byte array.

The argument structure is used for configuration flags as well as for reading and writing plugin parameters. It is a simple *variant type* structure which can represent various data types. The configuration flags can be set and read not only by the *processor script* to mark the current experimental configuration when a new record is obtained from hardware, but also by the plugins during the processing of a record. This way a plugin can evaluate a record and attach the result as a new flag to the record for another plugin to use during later processing.

The use of fixed length objects in the flag list helps to lower the CPU usage when copying, allocating and freeing records. With this design a record contains only two disjoint memory areas, one containing the actual record data and the other containing all flag values. This way the entire record content can be copied using two bulk memory copying calls which are implemented efficiently using SSE2²⁹ instructions. Additionally, this approach helps to minimize the memory fragmentation requiring only three memory allocations, one for the record itself and two for the flags and data. These normally are CPU intensive operations performed by the OS (operating system). To further reduce the CPU usage when handling records MOPS implements its own memory management mechanism for allocation and freeing of memory blocks. The memory is used by MOPS in very predictable patterns, as data arrays of identical size are used over and over again. The custom allocator maintains a map of free blocks and a map of currently used blocks. A freed block is not deallocated. Instead it is kept in the free map, sorted by size. When a new block is requested the map is checked and the smallest block that is greater or equal to the requested size is returned if available, else a new block will be allocated by the OS. When a memory block is provided by the OS it contains garbage data. Depending on the use case it may be necessary to initialize the block with 0's. This will require a processing thread to spend CPU time on. To avoid this MOPS clears all blocks in a dedicated thread after usage before returning them to the free map. Hence, the processing threads can spend all the available computing resources of their respective processor cores on processing the actual data. When a plugin allocates a memory block using the custom allocator, the plugin name is recorded. This way, if the application runs out of memory, it is easy to pinpoint which plugin caused the issue.

²⁹ Streaming SIMD (Single Instruction, Multiple Data) extensions

7.3. MOPS plugin interface

The interface for plugins is implemented using an *abstract class* and a *factory function* such that binary incompatibilities between compilers or different versions of the same compiler are circumvented. Each plugin is compiled as a library (DLL) exporting only one function:

```
extern "C" {
    API CPlugin* CreatePlugin(void* hPlugin, void* pInterface);
};
```

Code 7.3.1: Plugin factory declaration

This function takes two arguments: a pointer to the loaded library itself and an interface object. It returns a pointer to a newly created plugin instance, of the type `class CPlugin`, when successful, and `NULL` otherwise.

```
struct SInterface
{
    DWORD      Size;
    DWORD      Implementation;

    void(*QueueLogLine)(CPlugin* Plugin, UINT uFlags, const wchar_t* Line);

    size_t (*AvailMem)(void);
    void* (*Malloc)(size_t size, const wchar_t* strName);
    void* (*ReAlloc)(void *memblock, size_t size, const wchar_t* strName);
    void (*Free)(void *memblock);
    void* (*MemCpy)(void* dest, const void* src, size_t count);
    void* (*MemSet)(void* dest, int c, size_t count);

    void (*AppendFlags)(SFlag* &Flags, int &FlagCount);

    const wchar_t* (*GetInputPrefix)();
    const wchar_t* (*GetOutputPrefix)();

    const wchar_t* (*GetPluginName)(CPlugin* Plugin);
};
```

Code 7.3.2: Plugin Interface Structure

The interface object contains a `Size` field that is set to `sizeof(SInterface)` as implemented inside the core executable that is the total size in bytes of the interface structure, and a field called `Implementation` which contains the interface version. The rest of the structure holds function pointers. When a new function is added to the interface the size field will automatically increase. As long as neither a function is moved nor removed nor a function signature is changed the interface version should remain unchanged. An older plugin can operate with a newer interface object just fine as long as

it provides all known functions. Hence, the core application can be updated at any time, and more features added, without breaking binary compatibility and requiring all plugins to be updated. If it is chosen to break the binary compatibility by changing the interface structure, or the abstract plugin class, the interface version must be incremented. A plugin is deemed incompatible when the size of the interface object is smaller than the known size or when the version field is different from the expected value. When such a plugin is loaded accidentally it will know that the interface is incompatible and return `NULL` on the `CreatePlugin` call. This will log an error notifying the user of the incompatibility. If necessary in a broader deployment scenario plugins can be implemented in such a way that they could support more than one interface version.

The first function `QueueLogLine` allows a plugin to print text into the log window of the application. The first argument is a pointer to the plugin instance issuing the event, such that the core can retrieve the proper name of the plugin for the log display. The `uFlags` argument tells the log what color to use and whether the text should be printed in the *Dev-Log* only or also in the *User-Log*. The third argument is a pointer to the text as a 0 terminated string of 16 bit wide characters.

The next group of functions takes care of the memory management. Under windows it is common that each DLL links the “MSVC C runtime” statically, in such cases memory allocated in one application or library must not be freed in another. In MOPS, all memory used for storing records that is passed around between the plugins and the core is always allocated and freed by the core using an optimized custom allocator.

The current configuration state of the experiment is represented by flag values. The function `AppendFlags` takes a reference on a pointer to an array of such flags, and a reference on a flag counter, and adds all current flags to the array, incrementing the flag counter accordingly. If the pointer is `NULL` a new array will be created, else the existing array will be reallocated to the new size. In any case the pointer will be updated and other copies of the pointer will become invalid!

The following two functions return the paths to an optional input and output directory as pointers to 0 terminated wide strings. If a plugin needs to save data to disk or load data from disk it is expected to do that from the provided directories only.

The function `GetPluginName` was added later on. It allows a plugin to determine its own name as used inside the MOPS scripts. It is used for example to display the custom name

in the title of windows opened by the plugin. This comes in very handy if more than one instance of a particular plugin is used in an experiment profile.

```
void* g_hPlugin;
int g_Counter = 0;
SInterface theInterface;

CPlugin* CreatePlugin(void* hPlugin, void* pInterface)
{
    if(g_Counter == 0)
    {
        g_hPlugin = hPlugin;
        SInterface* Interface = (SInterface*)pInterface;
        if(Interface->Size < sizeof(SInterface) || Interface->Implementation != PLUGIN_IMPL)
            return NULL;
        memcpy(&theInterface, Interface, sizeof(SInterface));
    }
    g_Counter++;
    return new CSomePugin();
}
```

Code 7.3.4: The `CreatePlugin` function

When the function is invoked for the first time on a particular plugin it checks for compatibility and stores the interface object, and the pointer to the loaded library, in a global variable. On each call it increments the `g_Counter` variable by one, then it constructs a new instance of the plugin and returns it. The `g_Counter` variable indicates how many instances of the plugin are currently used. Every plugin class is derived from the abstract interface `class API CSomePlugin: public CPlugin:`

```
class CPlugin
{
public:
    virtual const wchar_t*    GetName() = 0;
    virtual const wchar_t*    GetInfo() = 0;

    /* ... a lot of code ... */

    virtual void*              Release() = 0;
};
```

Code 7.3.5: Abstract Plugin Class basics

The most basic functions of the interface are `GetName`, that provides a default name for the plugin, and `GetInfo` that provides a short description of the plugins function. These functions return pointers to constant 0 terminated wide strings, which are allocated statically inside the loaded library.

When the plugin instance is no longer needed it must be destroyed and freed using the `Release` function.

```

void* CSomePugin::Release()
{
    delete this;
    g Counter--;
    if(g Counter == 0)
        return g_hPlugin;
    return NULL;
}

```

Code 7.3.6: Release function

The `Release` function calls the destructor of the plugin instance and frees the memory, then it decrements the `g_Counter` variable. If the variable reaches 0 this means the last instance of the plugin just got destroyed and the plugin library can be unloaded from memory entirely. This is done by the core when the pointer to the loaded library is returned instead of `NULL`.

```

class CPlugin
{
    /* ... some code ... */

    virtual int      EnumPreset(int Preset, int& Enumerator) = 0;
    virtual int      GetPresetType(int Preset) = 0;
    virtual int      GetPresetName(int Preset, wchar_t* Name, int Size) = 0;
    virtual int      GetPresetOptions(int Preset, wchar_t* Options, int Size) = 0;
    virtual int      GetPreset(int Preset, wchar_t* Value, int Size) = 0;
    virtual bool      SetPreset(int Preset, const wchar_t* Value) = 0;

    virtual void      ShowDialog() = 0;

    virtual void      SavePresets(const wchar_t* FilePath, const wchar_t* Name) = 0;
    virtual void      LoadPresets(const wchar_t* FilePath, const wchar_t* Name) = 0;

    /* ... some more code ... */
};

```

Code 7.3.7: Abstract Plugin Class property access functions.

Each plugin is represented in the plugin lists by a root item with optional child items. The plugin preset sub-tree is created by querying the `EnumPreset` function starting with the argument `Preset` being `-1` and the argument `Enumerator`, which is passed by reference, being set to `0`. The function returns an integer value which is the identifier of the current entry and increments the `Enumerator` variable internally. The function is to be called in a loop until it returns `-1` indicating that no more entries are available. For each preset the function `GetPresetType` must be called to obtain the entry type which can be an actual preset or a sub-branch of presets. In the latter case for this entry a new loop has to be executed calling the `EnumPreset` function with the argument `Preset` being set to the value of the current entry identifier, with an own `Enumerator` variable initialized to `0`. Using

this recursive method a tree with an arbitrary amount of branches and sub branches can be constructed.

To get the tree entry name for the user interface `GetPresetName` is used. It takes the identifier as first argument, a pointer on a preloaded wide string and a size parameter telling the plugin the maximal string length that can be written. The returned parameter is actually the written string length. This mechanism ensures that the memory allocation is done by the calling function and is the default scheme for returning variable string values through the entire plugin interface. It will be referred to in the later text as an indirect string return.

Once the tree is constructed the preset values can be read using `GetPreset` taking the identifier as first argument and returning a string indirectly.

If the preset type as obtained from the `GetPresetType` call indicates the preset being a *combo box* the allowed values can be queried using `GetPresetOptions`, it indirectly returns a wide string containing all allowed values separated by "|". To write a preset there is `SetPreset` which takes the identifier and a pointer on a wide string with the new value. Some preset values may not be accepted by the plugin, the function returns true if the value was accepted and false otherwise.

Then there is the `ShowDialog()` function that is called when the configuration button in the plugin tree is pressed. This function is supposed to open a dialog where the user can change advanced settings that would not fit into the plugin tree, view information that cannot be natively presented in the measurement view, and interact with the plugin during runtime.

Finally, there are two functions to save and load the plugin configurations to and from an experiment profile. These functions take the path to the file and the custom name of the plugin, to identify an individual file section, such that multiple plugins of the same type can have different configurations.

```

[Scheduler]
'this section of the ini contains the Scheduler script

[Processor]
'this section of the ini contains the Processor script

[PluginName]
PluginParameter=123
AnotherPluginParameter=some string

[AnotherPluginName]
...

[YetAnotherPluginName]
...

[Scope]
ScopeLength=1723
ScopePos=0
ScopeHeight=10
ScopeOffset=0
SyncTo=
[Channel0]
Show=1
Location=PluginName.SomeStream>AnotherPluginName
ScopeOffset=-3.1
ScopeScale=0.5
BarOffset=0
[Channel2]
...

[ProcessorPlugins]
Plugin1=PluginName~PluginPath
...
Plugin*=AnotherPluginName~PluginPath

[SchedulerPlugins]
Plugin1=YetAnotherPluginName~PluginPath
...

```

Code 7.3.8: A profile file example.

The file format is a variation of a regular *.ini file, sections are named with in "[" "]" and contain one parameter per line starting with the parameter name, followed by a "=" and the rest of the line is the parameter value. However, there are two special sections that contain the configuration scripts as large multiline portions of plain text. There are also additional sections used for keeping other profile configurations which are regularly formatted. The *.ini *parser* and *sequencer* used by the plugins must not touch these sections. A plugin is supposed to use a section name containing at least its custom name as passed to the `SavePresets` and `LoadPresets` functions. In order to maintain access synchronization it is only allowed to write to the file when these functions are called.

```

class CPlugin
{
    /* ... some code ...*/

    virtual bool      StartPlugin() = 0;
    virtual void      AbortPlugin() = 0;
    virtual void      StopPlugin() = 0;
    virtual bool      IsRunning() = 0;

    // v1 Interface
    virtual int        GetParameter(const wchar_t* Name) = 0;
    virtual int        GetFunction(const wchar_t* Name) = 0;
    virtual bool       GetParameter(int Parameter, SArgument* Value) = 0;
    virtual bool       SetParameter(int Parameter, const SArgument* Value) = 0;
    virtual bool       CallFunction(int Function, const wchar_t* Arguments) = 0;
    //

    // v2 Interface
    virtual bool       GetParameter(const wchar_t* Name, SArgument* Value) = 0;
    virtual bool       SetParameter(const wchar_t* Name, const SArgument* Value) = 0;
    virtual bool       CallFunction(const wchar_t* Name, int iCount, SParam* pParams) = 0;
    //

    /* ... some more code ...*/
};

```

Code 7.3.9: Abstract Plugin Class scheduler script interface

When the core starts executing an experimental run, plugin operations are started using `StartPlugin`. Here, the plugin is supposed to initialize the hardware if it accesses any. The scheduler script can interact with the plugins by reading and writing parameters and by calling plugin functions.

There are multiple interface versions currently maintained for compatibility. The initial version identified each parameter and plugin function by an integer value internally, which first had to be retrieved for any given name string. The objective here was to translate all used names to integer identifiers during script compilation and use only jump tables during later execution. Hence, repetitive complex string comparisons could be avoided. However, this turned out to be insignificant in comparison to the overall script execution overhead, while slowing down rapid plugin development. Therefore, overwrite functions were provided that identify the parameters and functions directly by name and the caching mechanism is no longer used.

To read and write parameters `GetParameter` and `SetParameter` are used. They take a pointer on a wide string with the parameter name and a pointer on a `SArgument` object to which the parameter is written into or read from, respectively.

When the script calls a plugin function it uses `CallFunction` specifying the function's name as first argument, followed, depending on the interface version, either by a wide string containing an argument line, or by a parameter counter and an array of parameters.


```

struct SParam
{
    wchar_t    Name[32];
    DWORD      Size;
    wchar_t*    pData;
};

```

Code 7.3.10: Parameter structure

The parameter Structure used in the new `CallFunction` API contains a parameter name that can be up to 32 characters long, a size field containing the length of the data and a pointer on the data string itself. The structure must be allocated using the custom memory allocator such that the plugin can free the old data and attach a newly allocated data string. This way a plugin can return values back to the processor script directly.

When a parameter access function or the call function returns `false` this indicates an error and the measurement is aborted.

Once the measurement run is finished the plugin is stopped using `StopPlugin`. Then the plugin disconnects from hardware, if connected to any. This is done gracefully: any remaining operations will be finished before the function returns. If it is required to abort the experiment instantly `AbortPlugin` can drop all unfinished operations and terminate the plugin execution asynchronously.

```

class CPlugin
{
    /* ... some code ... */

    virtual int      GetStreamID(const wchar_t* Name) = 0;

    virtual int      EnumStream(int& Enumerator) = 0;
    virtual int      GetStreamName(int ID, wchar_t* Name, int Size) = 0;

    virtual bool     PullStream    (int ID, SSegment* &Segment, const wchar_t* Args) = 0;
    virtual bool     ProcessStream (int ID, SSegment* &Segment, const wchar_t* Args) = 0;
    virtual bool     PushStream    (int ID, SSegment* &Segment, const wchar_t* Args) = 0;

    virtual void     ClearStreams() = 0;

    /* ... some more code ... */
};

```

Code 7.3.11: Abstract Plugin Class processor script interface

The plugins process data records sequentially. Each processing sequence is executed in a different thread, and starts with a call to `PullStream`. This function takes an integer stream identifier, a reference on a pointer to a record object, and an optional string containing an argument line. When there is a record ready for processing the function sets

the record pointer to the new object, else the pointer is set to `NULL`. If the plugin determines that there will be no more records available it returns a *magic value* `END_OF_STREAM ((SSegment*)(-1))`. When a record value other than `NULL` has been obtained a loop is executed that iterates through all plugins specified for this sequence and calls `ProcessStream`. During the stream processing the plugin can update the record data, modify the flag list, and, if needed, drop the record entirely. In this case the pointer is set to `NULL` and the loop returns to begin prematurely. If the loop finishes it is checked whether a plugin was specified to take over the record. If this is the case this is done by calling `PushStream`, else the core frees the record itself and the whole sequence starts again. All three functions involved in record processing have the same argument list. The last argument containing an argument line is optional. It is evaluated only in the context of the currently handled stream and used to configure the handling function. For optimal performance it should only be evaluated once when the function is called for the first time during each run and the set configuration should be stored internally in a state associated with the current data stream.

Each plugin can handle multiple record streams. They are identified using unique integer values. In the processor script, however, each stream has a name. When the processor script is compiled all the names are mapped onto the unique values using `GetStreamID`, which takes the name as a pointer on a wide string and returns the identifier value. This way when the script is executed instead of string comparisons much more efficient jump tables or maps can be used. If there is no stream name specified in the script the default stream identifier is used which is simply `0`. A full list of streams can be retrieved using `EnumStream`. This function should be called in a loop with an `Enumerator` variable, which is passed by reference, initialized with `0`. On each call it will return a stream identifier and increment the `Enumerator` variable. Once all available stream identifiers have been returned it will return `-1`. At this point the loop should be terminated. To obtain the names of the streams for each identifier `GetStreamName` can be used, it indirectly returns the stream name. Plugins can be implemented to have a predetermined set of streams like, the Digitizer Card plugin which has exactly 4 hardware channels or such that they can process an arbitrary amount of different streams creating internal state sets for each newly encountered stream name.

To allow the users to preview the last information values produced for each stream, the stream states are not reset when the plugin finishes. Instead they are kept until the

function `ClearStreams` is called. This is done when a new experiment run is started before even calling `StartPlugin`.

```
class CPlugin
{
    /* ... some code ... */

    virtual int      EnumStatistic(int ID, int& Enumerator) = 0;
    virtual int      GetStatisticName(int ID, wchar_t* Name, int Size) = 0;
    virtual int      GetStatisticValue(int ID, wchar_t* Value, int Size) = 0;

    virtual int      GetScopeInfo(double x1, double x2, double y1, double y2,
                                   wchar_t* Info, int Size) = 0;

    /* ... some more code ... */
};
```

Code 7.3.12: Abstract Plugin Class info and stat interface

The plugins can provide information for the users, which are displayed in real time in the measurement view as a tree. The interface is implemented similarly to the plugin preset tree. The statistics sub trees are constructed recursively using `EnumStatistic` and `GetStatisticName`. However, there are no types to be queried. Each entry in the sub tree may therefore display a value that is to be read with `GetStatisticValue`, which indirectly returns the most recent value as a wide string.

The scope control located in the center of the measurement view can be used to obtain detailed information for any selected area using `GetScopeInfo`. The function is called with the coordinates of the selected rectangle and returns indirectly a string that is then displayed in the *User-Log*.

8. Publications

A universal matter-wave interferometer with optical ionization gratings in the time domain

Haslinger, P., Dörre, N., **Geyer, P.**, Rodewald, J., Nimmrichter, S. & Arndt, M.

Nature Physics. **9**, 144-148 (2013)

Photofragmentation beam splitters for matter-wave interferometry

Dörre, N., Rodewald, J., **Geyer, P.**, von Issendorff, B., Haslinger, P. & Arndt, M. ,

Physical Review Letters. **113**, 233001 (2014)

Refined model for Talbot-Lau matter-wave optics with pulsed photodepletion gratings

Dörre, N., Haslinger, P., Rodewald, J., **Geyer, P.** & Arndt, M.

Journal B: Optical Physics. **32**, 114-120 (2015)

A universal matter-wave interferometer with optical ionization gratings in the time domain

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Matter-wave interferometry with atoms [1] and molecules [2] has attracted a rapidly growing interest over the past two decades, both in demonstrations of fundamental quantum phenomena and in quantum-enhanced precision measurements. Such experiments exploit the non-classical superposition of two or more position and momentum states which are coherently split and rejoined to interfere [3–11]. Here, we present the experimental realization of a universal near-field interferometer built from three short-pulse single-photon ionization gratings [12, 13]. We observe quantum interference of fast molecular clusters, with a composite de Broglie wavelength as small as 275 fm. Optical ionization gratings are largely independent of the specific internal level structure and are therefore universally applicable to different kinds of nanoparticles, ranging from atoms to clusters, molecules and nanospheres. The interferometer is sensitive to fringe shifts as small as a few nanometers and yet robust against velocity-dependent phase shifts, since the gratings exist only for nanoseconds and form an interferometer in the time domain.

Recent progress in atom interferometry has been driven by the development of wide-angle beam splitters [14], large interferometer areas [15] and long coherence times [16]. Most interferometers operate in a Mach-Zehnder [5, 17], Ramsey-Bordé [18] or Talbot-Lau [19] configuration, some of them also in the time-domain [20, 21]. Here we ask how to generalize these achievements to atoms, molecules, clusters or nanoparticles - independent of their internal states.

Mechanical nanomasks [22] could be considered as universal if it were not for their van der Waals attraction on the traversing matter waves, which induces sizable dispersive, that is, velocity-dependent, phase shifts even for gratings as thin as 10 nm.

Optical [9, 14] or measurement-induced [23] gratings eliminate this effect, but most methods so far relied on closed transitions and required an individual light source for every specific kind of atom or molecule.

It is possible to circumvent this restriction by using the spatially periodic electric dipole potential in an off-resonant standing light wave. Its field then modulates the phase of the matter wave rather than the amplitude. This implies, however, that the spatial coherence of the incident matter wave needs to be prepared by other means before - such as by collimation, cooling [24] or the addition of another absorptive (material) mask [2].

Here, we demonstrate a new method for coherence experiments with a wide class of massive particles and show how a sequence of ionizing laser grating pulses [12] can form a generic matter-wave interferometer in the time-domain [13].

Figure 1 shows a schematic of the layout of our experiment, which we here realize specifically for clusters of anthracene (Ac) molecules. The molecules are evaporated in an Even-Lavie valve [25] that injects the organic vapor with a pulse width of about 30 μ s into

the vacuum chamber. The adiabatic co-expansion with a noble gas cools the molecules and fosters the formation of organic clusters - here typically up to Ac_{15} .

The bunch of neutral nanoparticles passes a differential pumping stage, enters the interferometer chamber and flies in a short distance (0.1 - 4 mm) from the surface of a super-polished CaF_2 mirror before it reaches the laser ionization region of a time-of-flight mass spectrometer (ToF-MS) where it creates the signal peaks.

The pulsed beams of three synchronized F_2 -excimer lasers ($\lambda = 157.63$ nm) hit the mirror surface and the cluster beam under normal incidence with a variable pulse energy of 1 - 3 mJ and a duration of about 7 ns. The laser beams are separated in space by ~ 20 mm along the cluster trajectory. Their mutual time delay is adjusted with an accuracy of a few nanoseconds. We choose the laser beam diameters (~ 1 mm $\times$ 10 mm rectangular flat top, extended along the cluster beam) to cover a wide particle bunch emitted by the source, whereas the detection laser beam is narrow enough to post-select only those clusters that have interacted with all three laser light pulses.

All three laser gratings interact with the matter waves in two different ways [13]: they imprint a periodic phase and, more importantly, they act as transmission gratings because the photon energy of ~ 7.9 eV exceeds the ionization energy of the nanoclusters. Particles that traverse the antinodes of a laser grating ionize with high probability after absorption of one or more photons and a weak electric field removes them from the beam. Close to the nodes of the standing light waves the clusters remain neutral and move on in the interferometer. This process imprints a periodic modulation onto the matter-wave amplitude - as if the clusters had passed a mechanical nanomask.

A strong spatial localization inside the first laser grating is important for preparing a comb of emergent wavelets whose transverse coherence will cover a few antinodes in the second light grating further downstream. This is a prerequisite for interference to occur, that is, for

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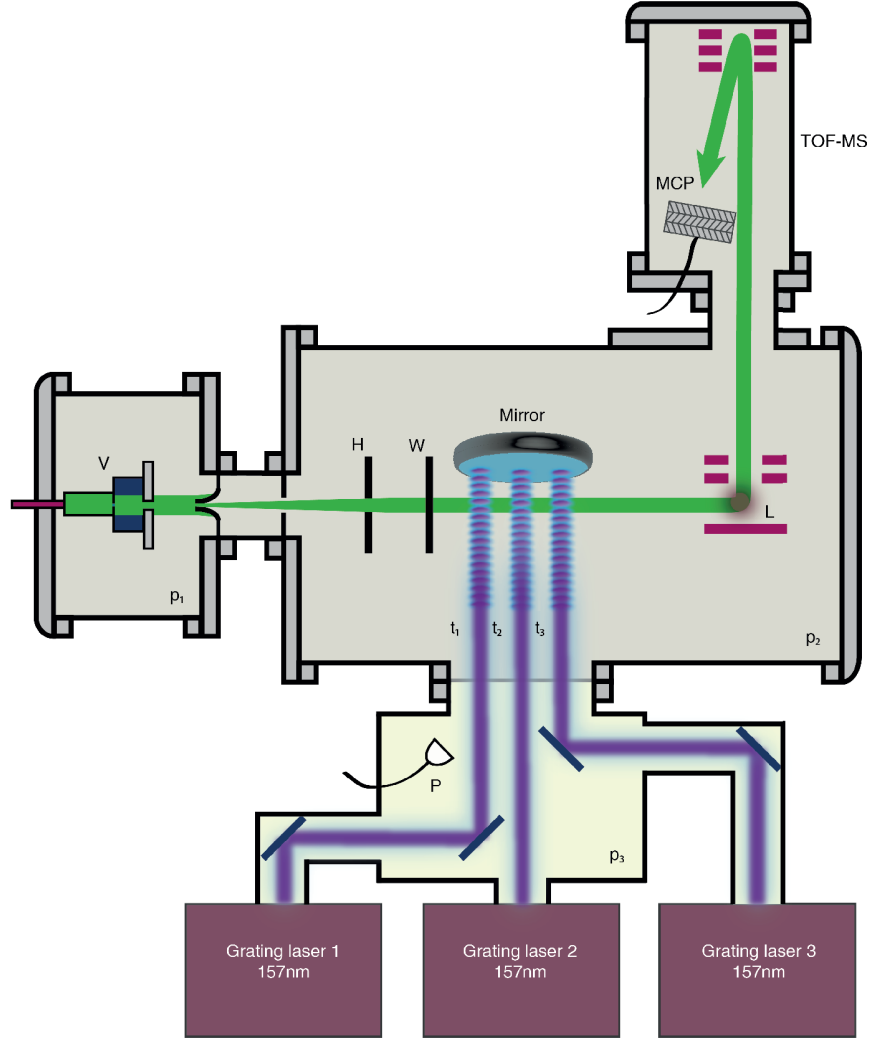


FIG. 1. **Layout of the OTIMA interferometer.** (a) Setup for nanoparticle interferometry with three short-pulse optical ionization gratings. From left to right: The Even-Lavie valve (V) produces a $30 \mu\text{s}$ pulse of neutral anthracene clusters which are cooled in an adiabatic co-expansion with a noble gas jet. The cluster beam is delimited by two slits which are variable in height (H) and width (W). The laser pulses at $t_1 = 0$, $t_2 = T$ and $t_3 = 2T$ are back-reflected by a single two-inch mirror to form three standing light waves. These are responsible for preparing the initial spatial coherence, for matter-wave diffraction and for spatially filtering the emerging cluster interferogram. The detection laser (L) ionizes the transmitted neutral clusters for time-of-flight mass spectrometry (TOF-MS). A photodiode (P) is used to monitor the laser timing with nanosecond accuracy. (b) The interferogram is formed by multiple paths from the first to the third grating which correspond to an effective momentum transfer of $n\hbar k$ in each grating, with $n \in \mathbb{Z}$. Accurate timing ensures that the interfering paths branch and close at the same points on the grating axis x , irrespective of the cluster's initial velocities v_1 (red) $>$ v_2 (green). The stars indicate the localization of the matter waves.

the formation of a free-flying cluster density pattern at precisely defined moments in time, which is probed with nanosecond precision by the third ionizing standing wave.

The three laser pulses form a Talbot-Lau interferometer in the time domain, which exhibits transmission resonances when the delay between two subsequent pulses is close to the Talbot time $T_m = m d^2 / \hbar$, with m the cluster mass and $\hbar$ Planck's constant. In our setting the grating period $d = \lambda / 2 = 78.8 \text{ nm}$ results in $T_m = 15 \text{ ns/amu}$. All particles

see the same gratings at the same time irrespective of their velocity. Even though they may enclose different areas in real-space ($x - z$), they will accumulate the same phase and contribute constructively to the same interferogram for each given mass (Figure 1b).

We trace the emergent interference pattern in four different ways: its mass characteristics, its dependence on the pulse separation and pulse sequence asymmetry, and by visualizing its structure in position space.

We start by monitoring the ToF-MS signal and toggle

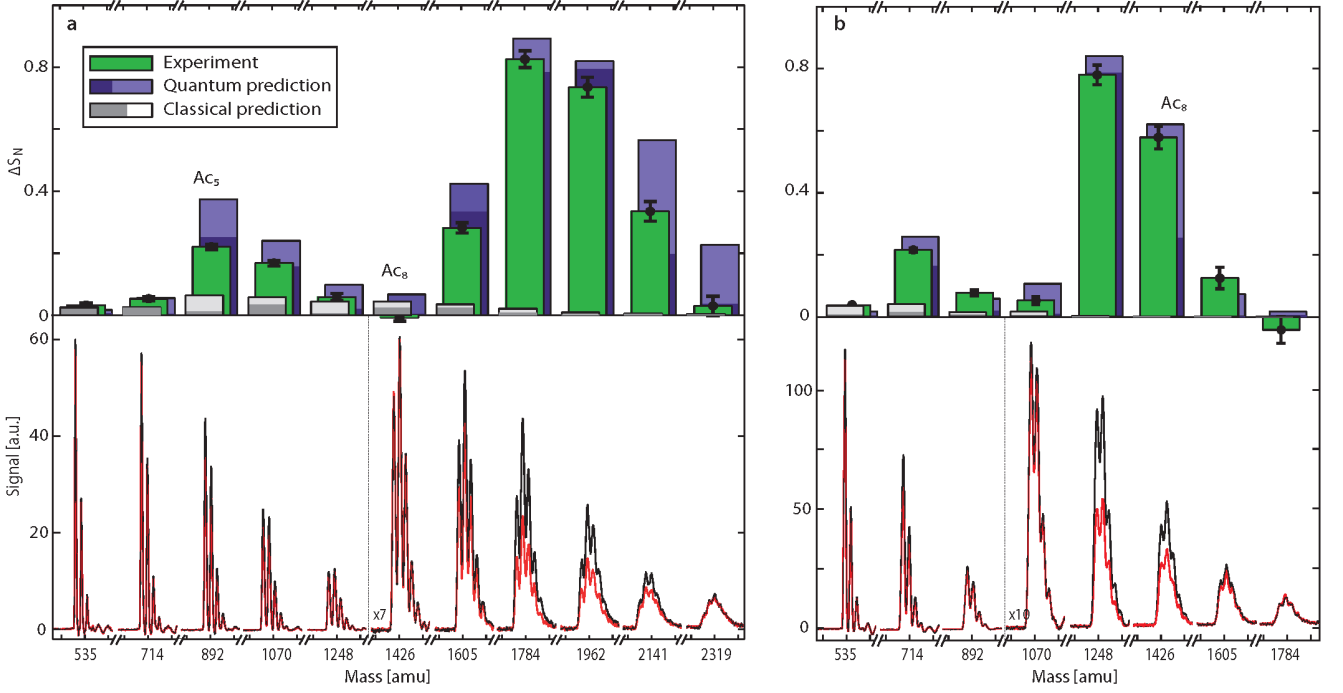


FIG. 2. **Cluster interference visualized via the mass spectrum, for two pulse separation times.** (a) Bottom panel: Mass spectra recorded for a resonant (black line) and off-resonant ($\Delta T = 200$ ns, red line) pulse separation of $T = 25.9$ μ s (clusters seeded in an argon jet). Each cluster signal splits in isotopic sub-peaks. The x-ticks correspond to a mass separation of 4 amu. The two spectra differ for masses which fulfill $T_m \simeq T$. Top panel: Histogram of the cluster interference contrast, as measured by the signal difference ΔS_N integrated over the main isotopes of a given cluster. The predictions of the quantum/classical model [13] are shown in violet/grey. The light violet/grey regions indicate the variation of the fringe contrast with a $\pm 30\%$ variation of the cluster polarizability α_{157} . For further details, see Methods and Suppl. Inf. e (b) Same as (a) but with neon seeding and $T = 18.9$ μ s. The error bars represent one standard deviation of statistical error (see Suppl. Inf. h).

between a resonant and a non-resonant setting. In the resonant mode the delays $t_2 - t_1 = T$, $t_3 - t_2 = T + \Delta T$ between two subsequent laser pulses are equal, $\Delta T = 0$, and quantum interference is expected to modulate (enhance or reduce, depending on the phase) the transmission for the mass whose Talbot time matches the pulse separation T . In the off-resonant mode, the pulse delays are imbalanced by $\Delta T = 200$ ns and this tiny mismatch suffices to destroy the interferometric signal. We extract the interference contrast from the normalized difference $\Delta S_N = (S_R - S_O)/S_O$ between the resonant S_R and the off-resonant signal S_O and plot it as a function of mass in Figure 2. The experimental mass spectra and ΔS_N bars (green) can be well understood by a quantum mechanical model (violet bars), as described in the Methods Section, and both are in marked discrepancy with a classical model (grey bars) [13].

The role of the pulse separation T is demonstrated by changing the seed gas from argon to neon. Shifting the most probable jet velocity from 690 to 925 ms^{-1} allows us to decrease T . The quantum model then predicts the highest contrast to occur at smaller masses, as confirmed by the experimental data in Figure 2b.

Figure 3 shows a clear resonance in ΔS_N as a function

of the time imbalance $\Delta T \in [-70, +70]$ ns with a width determined by the transverse momentum distribution of the cluster beam [13]. The momentum spread inferred from a Gaussian fit to the data in Figure 3 corresponds to a divergence angle along the grating of 2.1 mrad, in good agreement with the experimental settings.

In our set-up, the pulsed supersonic expansion determines the cluster velocity distribution and the pulsed mass detection post-selects its relative width to $\Delta v/v \simeq 3\%$. It is then justified to interpret the observations in position space: With the de Broglie wavelength given by $\lambda_{dB} = h/mv$ the mass distribution also represents a wavelength spectrum. The most prominent interference peak in Figure 2b at 1248 amu corresponds to the heptamer Ac_7 with $\lambda_{dB} \simeq 345$ fm, at $v \simeq 925$ ms^{-1} . The highest mass peaks in the spectrum reach down to below $\lambda_{dB} \simeq 275$ fm.

Finally, we can also prove the formation of an interference pattern in real space by modifying the period of the central grating: While all laser beams had originally been set to normal incidence on the interferometer mirror - with an uncertainty of about 200 μ rad - we now explicitly tilt the central laser beam by 5.1 mrad along the cluster beam. The direction

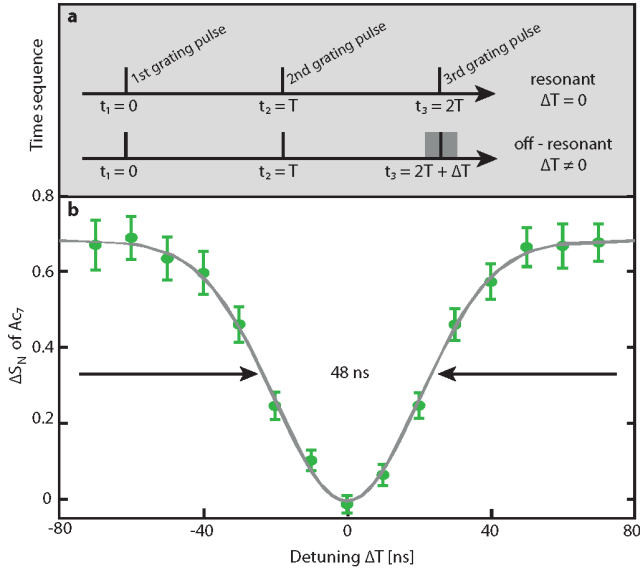


FIG. 3. **Interferometric resonance and timing precision.** Cluster self-imaging in a pulsed near-field interferometer is a resonant process with a short acceptance window for the matter waves to rephase. (a) Pulse sequence and (b) difference ΔS_N between the resonant and off-resonant signals detected at a mass of Ac_7 as a function of ΔT . In our setup and for a pulse separation time T of $18.9 \mu s$ interference occurs during a time window of 48 ns (FWHM). The error bars represent one standard deviation of statistical error (see Suppl. Inf. h).

of the standing-light-wave grating remains defined by the orientation of the mirror surface, but an increasing tilt angle θ reduces the modulus of the wave vector perpendicular to the surface, $k_p \simeq k \cdot \cos \theta$. We can shift the interference pattern by half a grating period when the clusters pass the mirror surface at an average distance of 1.5 mm. We plot the fringe shift as a function of the separation between the beam and the mirror in Figure 4 and find a damped sinusoidal transmission curve for all clusters with the expected period. The overall damping results from the limited coherence of the laser system and the vertical extension of the Ac cluster beam.

All tests presented here confirm the successful experimental realization of an optical time-domain ionizing matter-wave (OTIMA) interferometer [2, 13], which exploits pulsed ionization gratings. This versatile tool for quantum interferometry will be applicable to a large class of nanoparticles. Owing to the pulsed gratings, all phase shifts caused by constant external forces become velocity-independent and leave the contrast unaffected. The dispersive Coriolis shift [15] can be well compensated by a suitable orientation of the interferometer, if needed.

The wide applicability and non-dispersive nature of pulsed ionization gratings make the OTIMA

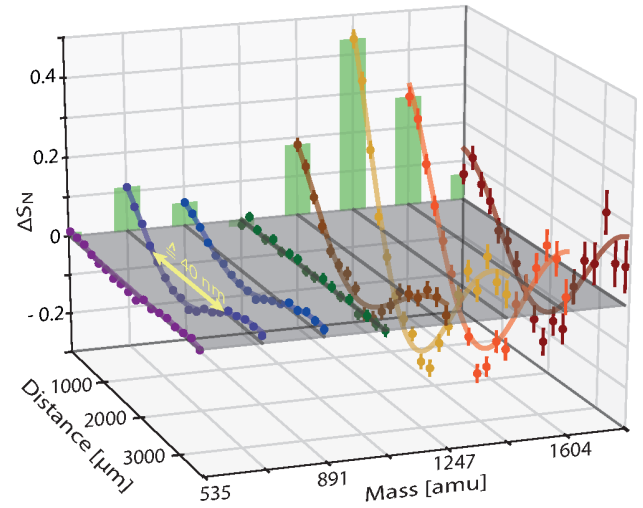


FIG. 4. ΔS_N as a function of the mirror displacement for different clusters. The second grating laser beam was tilted by 5.1 ± 0.3 mrad in the direction of the molecular beam to stretch the effective grating period by about 0.013 per mille. This suffices to induce a fringe shift of half a grating period for molecules travelling around 1.5 mm distance from the mirror surface. The mirror height is varied to effectively shift the second grating with regard to the other two which allows us to scan the cluster interference pattern. We extract the periodicity for ΔS_N as a function of the mirror distance by fitting a damped sine curve to the experimental data. This periodicity corresponds to the expected effective period [13] of the interferogram of 80 nm. The error bars represent one standard deviation of statistical error (see Suppl. Inf. h).

design particularly appealing for quantum experiments with highly complex particles, eventually even with nanoparticles at the length scale of the grating period. As high-mass interferometry requires coherence of the order of the Talbot time, practical mass limits are imposed by free fall in the gravitational field on Earth in combination with the limited coherence of vacuum ultraviolet lasers and the finite phase-space density of the available particle sources. However, none of them is fundamental. Even in the presence of thermal radiation at room temperature (particle and environment) and collisional decoherence at a background pressure of 10^{-9} mbar, the OTIMA design is predicted to enable new tests of quantum physics, such as tests of spontaneous localization, with particle masses around 10^6 amu and beyond [26].

On the applied side, the OTIMA set-up is expected to improve the accuracy of molecule and cluster deflectometry because it ensures the same interaction (phase accumulation) time for all particles with the external fields [27] and a position readout at the nanometre scale. Our interferometer concept therefore establishes also the basis for a new class of quantum-enhanced precision metrology experiments.

METHODS

Absorption and optical polarizability. The central grating influences the propagation of the coherent matter wave by modulating both its amplitude and phase. It does this by removing particles from the anti-nodes of the standing light field and by imprinting a phase onto the matter wave in proportion to the clusters' optical polarizability at 157 nm. In the first and third grating the phase modulation has no effect, since the clusters enter with random phases, and since the last grating merely acts as a transmission mask. Neither the absorption cross sections $\sigma_{157}(N)$ nor the polarizabilities $\alpha_{157}(N)$ are known, a priori, for each cluster of N molecules in the vacuum ultraviolet wavelength range. However, $\sigma_{157}(N)$ enters the model only through the mean number of photons absorbed $n_0(N)$ in each grating which we can determine by monitoring the cluster loss rate. While this parameter influences the general shape of the interference curve as a function of mass, the polarizability may modify the predicted contrast of each individual cluster. We assume the polarizability and the absorption cross section to exhibit the same N -scaling as retrieved from our $n_0(N)$ measurements and we allow the polarizability to vary by $\pm 30\%$ (light violet confidence areas in Figure 2) around the single-molecule value. We use the polarizability

$\alpha_{157}(1) = 25.4 \times 10^{-30} \text{ m}^3$ from Marchese et al. [28] and we extract an absorption cross section of $\sigma_{157}(1) = 1.1 \times 10^{-20} \text{ m}^2$ from Mallocci et al. [29].

This yields the quantum and classical theory curve in Figure 2. Apart from the uncertain polarizability, the deviations from the experimental data may be attributed to a limited efficiency of single-photon ionization and contributions by fragmentation processes. While the absolute interference contrast is sensitive to a variety of different cluster properties which still wait to be extracted in combination with more refined cluster theory, the fringe shift will become valuable for precisely measuring the interplay between internal cluster properties and external forces.

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SUPPLEMENTARY INFORMATION

(a) Source

We use an Even-Lavie (EL) valve to create a pulsed neutral molecular cluster beam. Anthracene (Ac) molecules are heated close to their melting point (491 K) in the valve and they are co-expanded into high vacuum with a supersonic noble gas jet ($p \simeq 1 - 10$ bar). There

they cool and condense to form clusters ranging from $Ac_1 - Ac_{15}$. The EL valve is operated at a repetition rate of 100 Hz and it is synchronized with the three vacuum ultraviolet (VUV) grating lasers and the detection laser.

(b) VUV Laser system

The gratings are generated by three synchronized GAM lasers, model EX50F, $\simeq 5$ mJ, shot-to-shot energy stability $\simeq 5\%$, coherence length $\simeq 1$ cm. The grating transmission function depends on the laser energy which we monitor for every individual pulse via the photodiode P (in Figure 1). All mass spectra are sorted according to a given laser energy and pulse delay ΔT . The laser beam lines are evacuated and purged with dry nitrogen (6.0) at 1 mbar to avoid both absorption and laser-induced deposition of debris on the mirrors. The ionizing VUV laser at the TOF-MS is a Coherent Excistar F₂-laser (3 mJ) with an energy stability of better than 5%.

The F₂ laser operates mainly at 157.63 nm, with an additional weak line at 157.52 nm [30]. Our own measurements on the GAM lasers confirm the specified coherence length of $l_c \simeq 1$ cm which ensures a standing wave in a few millimeters distance to the mirror. The transverse coherence of each excimer laser is given by its output aperture and amounts to $\simeq 40 \mu\text{m}$ at the mirror.

The timing sensitivity of the OTIMA scheme at short pulse separation periods requires a precise monitoring of the laser jitter. The intrinsic short term jitter of all three GAM lasers is less than 7 ns (FWHM). They exhibit, however, long term instability of the order of 100 ns, which we measure and compensate. Our Coherent laser jitters by $\simeq 20$ ns (FWHM). The timing of the grating laser pulses is recorded to post-select the interferograms according to their pulse-delays. All measurements were made with a maximal jitter smaller than 5 ns.

(c) VUV mirror

The dielectric interferometer mirror (Jenoptik, Germany) is made from VUV grade CaF₂ coated with a reflectivity of $R > 96\%$ under normal incidence. The finite reflectivity allows us to monitor the position and shape of the laser beams via their scattering on the frosted backside of the mirror.

Moreover it causes a small running wave to add to the standing wave. The constant intensity offset would only slightly reduce the interference contrast if every cluster was always ionized by a single photon. Our own and independent measurements indicate a minimal spherical deformation across the two-inch mirror towards its edges up to 100 nm.

(d) Mass spectrometry

The time of flight mass spectrometer (ToF-MS, Kaesdorf Munich) is built as an orthogonal reflection MS with $\Delta m/m = 1/3000$. The relative mass spread across every individual multiplet is as small as 0.1 - 1 %. A mass variation of ± 2 amu on $m = 1400$ amu gives rise to a variation of 30 ns on 20 μ s Talbot time. This leads only to a negligible reduction of the interference contrast.

(e) Number of absorbed photons

We chose anthracene as a test molecule because its ionization energy E_i is smaller than the photon energy at 157.63 nm (7.9 eV) and the contrast is highest if the absorption of a single photon suffices to ionize the particle. If this condition is fulfilled for a certain cluster number N it will be generally met for all higher clusters too, since E_i decreases with cluster size to approach the work function of the bulk. Photon absorption without subsequent ionization would diminish the interference contrast. The photoionization quantum yield [31] of anthracene at 7.9 eV is only 10 %. Our data are compatible with the assumption that it is close to one for clusters composed of several molecules. Different structural isomers may respond differently to the incident light, but a full assessment of all optical properties for all cluster sizes is beyond the scope of this first demonstration of experimental OTIMA interferometry.

(f) Vacuum system

The source chamber is evacuated to $p_1 = 1 \times 10^{-5}$ mbar, the interference chamber to $p_2 = 2 \times 10^{-8}$ mbar and the optical beam line to $p_3 = 1$ mbar.

(g) Data Recording and Processing

The TOF-MS voltage signal is recorded using a 10 bit digitizer card (Agilent Acquiris DC282) with 0.5 ns time resolution. We run the experiment with 100 Hz repetition rate. A data file for one mass spectrum has a size of 1

mega points. Data are post-processed in real time using a custom developed software solution. The software also records the laser timings and pulse energies.

(h) Figures

Figure 2: The TOF-MS data were averaged over about 28000 individual mass spectra for panel (a) and about 14500 spectra for panel (b). An overall TOF-MS background was subtracted, for all masses equally. The green columns in the upper panels of Figure 2a and 2b were generated by summing the mass spectra (bottom panels) over a mass region whose width is indicated by the width of the columns. It accounts for the majority of the isotopic spread of a given cluster peak. The experimental error bar was determined as follows: Since the experimental response to the incidence of an ion is a voltage peak whose amplitude changes both from shot to shot and with increasing ion mass, we chose to extract a measure for the true count rate from the observation of “no count” - a small discriminator threshold was set to distinguish between the presence or absence of ions - in every mass bin. Assuming a Poissonian distribution of the cluster counts one can then infer the average detected cluster number and its standard deviation (δS_O and δS_R) from the probability of finding zero counts. The error of the normalized signal difference $\delta(\Delta S_N)$ is then computed using Gaussian error propagation. The data has been evaluated and plotted with Matlab R2010b and arranged using Adobe Illustrator CS5.

Figure 3: For this data set, TOF-MS data were averaged over 3300 - 3500 frames for each data point. The error bar was determined by the same procedure as in Figure 2. The data has been evaluated and plotted with Matlab R2010b and arranged using Adobe Illustrator CS5.

Figure 4: For this data set we averaged over roughly 25000 mass spectra for every step in mirror distance. Uncertainty bars were generated using the same procedure as in Figure 2 and 3. The data has been evaluated with Matlab R2010b, plotted using the Matplotlib package for Python and arranged using Adobe Illustrator CS5.

Photofragmentation Beam Splitters for Matter-Wave Interferometry

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Extending the range of quantum interferometry to a wider class of composite nanoparticles requires new tools to diffract matter waves. Recently, pulsed photoionization light gratings have demonstrated their suitability for high mass matter-wave physics. Here we extend quantum interference experiments to a new class of particles by introducing photofragmentation beam splitters into time-domain matter-wave interferometry. We present data that demonstrate this coherent beam splitting mechanism with clusters of hexafluorobenzene and we show single-photon depletion gratings based both on fragmentation and ionization for clusters of vanillin. We propose that photofragmentation gratings can act on a large set of van der Waals clusters and biomolecules which are thermally unstable and often resilient to single-photon ionization.

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Recent explorations of matter-wave physics with very massive particles [1] have been motivated by the rising interest in new tests of the quantum superposition principle [2–5] and quantum sensors. This has triggered the question which scheme might be best adapted to diffract complex nanomatter in a coherent way. Earlier experiments with absorptive masks of light were based on the possibility to prepare dark states in atoms [6, 7]. The manipulation of composite particles requires, however, mechanisms which are largely independent of internal particle properties or particular resonances. Matter-wave interferometry with optical absorption gratings in the time domain (OTIMA) has recently been demonstrated with clusters of anthracene molecules [8]. This scheme [9, 10] is scalable to high masses and has been realized for materials that can be ionized by a single photon [11, 12] of energy 7.9 eV. This vacuum ultraviolet (VUV) light can be coherently generated by commercially available fluorine excimer lasers. However, the ionization energy of many organic or biological molecules exceeds 8 eV and is too high for single-photon ionization gratings.

Here we show that the thermal instability of composite particles, which is often a hindrance in physical chemistry and quantum optics experiments, can be exploited to realize a coherent beam splitter for complex matter. We demonstrate specifically how single-photon absorption in the antinodes of a standing light wave can lead to particle heating and fragmentation and, therefore, to a spatially periodic depletion of the cluster beam. Each light grating acts similarly to a mechanical mask and functions as a diffraction element. The light pulses trigger the depletion and form together an interferometer in the time domain. What counts is the act of measurement in each grating, which labels a periodic set of particles in the beam, as “to fragment before detection”. All particles carrying the complementary property “nonfragmented” are then read and registered by the detector.

The experimental setup (see Fig. 1) has been described

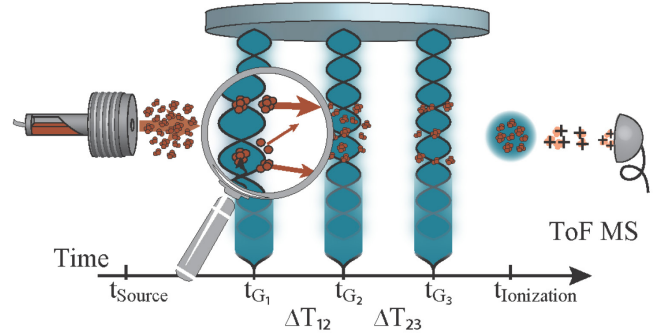


FIG. 1. Time-domain interference using single-photon fragmentation gratings. A 500 K pulsed nozzle source emits organic molecules, here hexafluorobenzene (HFB) or vanillin. Supersonic expansion in an intense neon pulse leads to the formation of clusters. Three standing light-wave gratings form the matter-wave interferometer. At the antinodes of the light gratings, the clusters may fragment or ionize after absorption of a single 7.9 eV (VUV) photon. This leads to a pulsed and spatially periodic labeling of clusters and their effective removal from the beam. Only clusters transmitted through the absorptive light comb contribute to the interference pattern.

in [8, 9]. Molecules are evaporated and emitted by a pulsed (20 μ s, 100 Hz) Even-Lavie valve [13] to form van der Waals clusters during adiabatic cooling in a coexpanding noble seed gas. The particle cloud passes in close proximity to a two inch dielectric mirror where it is subjected to three VUV laser light pulses (7 ns, $\lambda_L = 157.63$ nm, 3 mJ in 1×10 mm²). The light forms standing waves upon retroreflection at the mirror surface. In order to impose spatial matter-wave coherence onto the incident cluster beam, the first grating pulse G_1 must be absorptive; i.e., particles in the antinodes must be removed from the detected signal with high efficiency. The node regions then act as sources for elementary matter wavelets. If these sources are sufficiently small, the emerging waves will expand coherently to overlap several nodes and antinodes in the second grating G_2 . A cluster

density pattern forms by virtue of the Talbot-Lau effect as a self-image of G_2 which is sampled by the absorptive third grating G_3 [1, 9].

While the interference contrast is only determined by the absorptive (depleting) character of the cluster-light interaction in G_1 and G_3 , we also need to consider the dipole interaction between the laser light field and the cluster's optical polarizability in G_2 . This coupling imprints a spatially periodic phase onto the matter wave in addition to the amplitude modulation that is caused by depletion [8]. The particles that are transmitted through the interferometer are ionized by 157.63 nm light (10 ns, 0.2 mJ in $1 \times 3 \text{ mm}^2$) and analyzed in a time-of-flight mass spectrometer (ToF-MS). For interference measurements, the power in each grating is adjusted such that less than 25% of the particles are transmitted. This determines the opening fraction of the grating. The pulse energy of the center grating can be attenuated *in situ* using a 10 mm long pressure cell which allows us to vary the amount of air in a segment of the evacuated beam line. Since oxygen strongly absorbs in the VUV [14], a variation of the air pressure inside the cell between 10^{-4} and 200 mbar is sufficient to reduce the incident laser energy from 90 to almost 0%. In order to monitor pulse-to-pulse variations of the laser power, we use GaP photodiodes to record the relative power of all laser pulses which we cross correlate with the detected ion signal.

The three grating laser pulses form a time-domain Talbot-Lau interferometer if the delays between two pulses are equal [8]. This pulse separation time is related to the interfering mass m via the Talbot time $T_T = md^2/h$, where $d = \lambda_L/2$ is the grating period and h is Planck's constant. Matter-wave interference can then be seen in the intensity modulation of the mass spectrum (see Fig. 2) [8]. The signal is measured in two complementary modes: an interference mode (S_{Int}) in which the grating pulse separation times are equal, $\Delta T_{12} = \Delta T_{23}$ (Fig. 1), and a reference mode (S_{Ref}) in which the two times differ by several tens of nanoseconds, $\Delta T_{12} = \Delta T_{23} + \Delta T$, so that no matter-wave interference can be measured. This is used to express the visibility of the interference pattern in terms of the normalized signal contrast $S_N = (S_{\text{Int}} - S_{\text{Ref}})/S_{\text{Ref}}$.

We use clusters of hexafluorobenzene ($m=186 \text{ u}$ per monomer) and vanillin ($m=152 \text{ u}$ per monomer) as examples for nanoparticles with ionization energies above or close to the grating's photon energy. The vertical ionization energy of hexafluorobenzene (HFB) and vanillin monomers are 9.97 eV [15] and 8.30 eV [16], respectively. Although the ionization potential may fall with increasing cluster size, measurements on benzene indicate that for organic clusters it will not fall by more than 10% below the value of the monomer [17].

Single-photon ionization is energetically excluded for small HFB and vanillin clusters. Nevertheless, we observe a substantial interference contrast S_N as a function

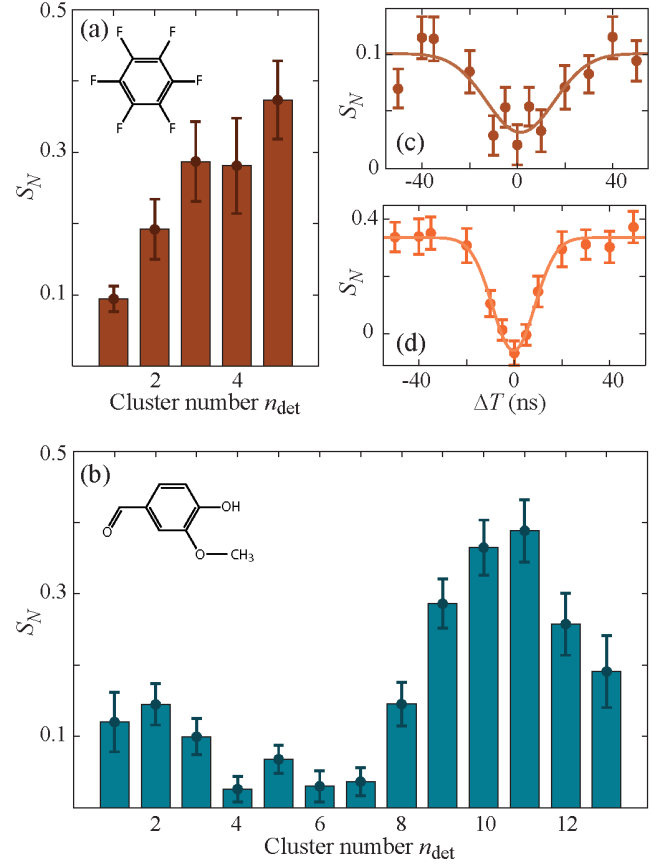


FIG. 2. Quantum interference of clusters of hexafluorobenzene (a) and vanillin (b). We monitor the normalized signal contrast (S_N) which compares the cluster transmission for the on resonant and off resonant setting of the grating pulse separation times. Resonances can be seen in the mass spectrum when the pulse separation time is close to an integer multiple of the Talbot time T_T . (c, d) The resonant character of quantum interference can be seen by varying the difference of the two pulse separation times $\Delta T = \Delta T_{12} - \Delta T_{23}$ between subsequent diffraction gratings in the reference mode. Interference occurs only when both times are equal. A temporal detuning of several dozen nanoseconds suffices to destroy the effect. The dips in (c) and (d) were measured for the detected cluster number $n_{\text{det}} = 1$ and $n_{\text{det}} = 5$ of HFB. The error bars represent one standard deviation of statistical error. The solid lines are Gaussian fits.

of the detected cluster mass for both species (Fig. 2). The separation time between the gratings was set to $11.5 \mu\text{s}$ and $18.9 \mu\text{s}$, respectively, which corresponds to the Talbot time of the fourfold cluster of HFB and the eightfold cluster of vanillin. For HFB, we have also measured the temporal width of the interference resonance [Figs. 2(c) and 2(d)]. As expected for time-domain Talbot-Lau interference [8], high contrast is only observed if the delay between two grating pulses is equal to within a few nanoseconds. In this case, the interference signal and the reference signal are identical and S_N vanishes.

Demonstrating fragmentation as the cause of the beam splitting process is challenging since the depletion mechanism leaves no trace in the final interference pattern. Ideally, the detector records only those particles that have *not* absorbed a photon in any of the gratings. In addition, the beam splitting angle and the momentum transfer to the particles depend only on the grating geometry and the particle polarizability. Clusters that absorb a photon may either ionize (followed by extraction from the beam with an external electric field) or fragment. The fragments are unaffected by the field; however, assuming evaporation in thermal equilibrium, the cluster fragments will reach an escape velocity beyond 100 ms^{-1} . At a forward cluster velocity of 900 ms^{-1} , the majority of all parent clusters and molecules are therefore ejected beyond the detector acceptance angle of 10 mrad .

In order to corroborate the beam splitter mechanism, we first show that photoionization requires at least two photons while depletion in the light gratings is a single-photon effect. For that purpose, we have recorded the cluster intensity as a function of the detection laser energy E_D as sketched in Fig. 3(a). For HFB, we observe a strongly nonlinear power dependence in Fig. 3(b) for all detected clusters at low laser energy consistent with a resonantly enhanced single-photon absorption cross section at 157 nm [18] and a multiphoton ionization process [19]. The detected cluster distribution $S(n_{\text{det}})$ must therefore differ from the incident cluster distribution $S(n_{\text{inc}})$ since fragmentation in the ionization stage depletes larger clusters and replenishes the signal intensity at smaller cluster numbers.

For small clusters of vanillin, we also observe a nonlinear power dependence which gradually changes to a linear one-photon behavior for larger clusters [Fig. 3(c)]. We attribute this transition to the small difference between the photon energy (7.9 eV) and the ionization energy of the vanillin monomer (8.3 eV), which will be further reduced for large clusters. In the limit of small laser energy E_D , the signal can be expanded to second order: $S_{\text{ion}} \sim AE_D + BE_D^2$. For small clusters the power series is dominated by the quadratic term ($A=0$) whereas for $n > 3$, the emergence of a linear component indicates a one-photon contribution, too.

If the matter-wave beam splitters were dominated by multiphoton processes, we should see a similar nonlinear dependence in the reduction of the cluster transmission as a function of the laser energy E_G in G_2 . In order to compare ionization and transmission data, we have reduced the grating to two counter propagating running waves by shifting the interferometer mirror beyond the coherence length of the grating laser [Fig. 3(d)]. The observed beam depletion in Fig. 3(e) (HFB) and Fig. 3(f) (vanillin) is now well represented by exponential curves for all cluster numbers n_{det} . This is expected for a single-photon depletion process with Poissonian statistics. Since ionization was shown to require at least two photons for

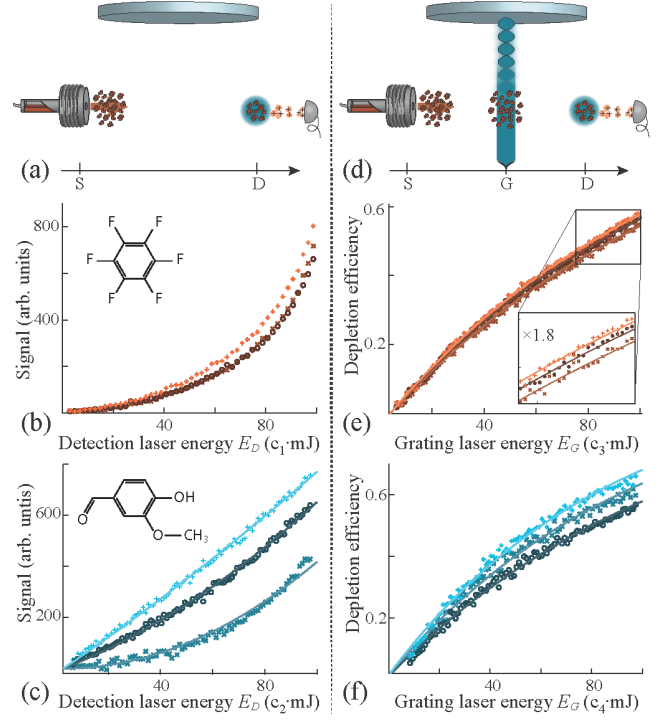


FIG. 3. (a) Verifying the multiphoton character of the cluster ionization process: clusters propagate freely between the source S and the detector D where they are ionized upon photoabsorption. (b) Photoionization of HFB clusters: the detected ion signal depends nonlinearly on the laser energy E_D . This is a clear sign of a multiphoton process. The curves represent $n_{\text{det}} = 2$ ($\times$, orange), 4 ($\circ$, dark orange), 5 ($+$, light orange) HFB molecules per detected cluster. (c) Photoionization of vanillin clusters: $n_{\text{det}} = 3$ ($\times$, blue), 7 ($\circ$, dark blue), 11 ($+$, light blue). We observe a high-order nonlinearity for $n_{\text{det}} = 3$ and an indication of single-photon events at $n_{\text{det}} = 11$. This is consistent with the expectation that the ionization efficiency increases with increasing cluster number. (d) Verifying the single-photon character of the beam depletion process in the laser grating G : The mirror has been retracted beyond the coherence length of the F_2 laser to limit the cluster-light interaction to absorption in a running wave. (e) Beam depletion, case of HFB: the curves are well reproduced by single-photon events in a Poissonian process (exponential fits) for the detected cluster numbers $n_{\text{det}} = 2$ ($\times$, orange), 4 ($\circ$, dark orange), 5 ($+$, light orange). (f) Beam depletion, case of vanillin: $n_{\text{det}} = 3$ ($\times$, blue), 7 ($\circ$, dark blue), 11 ($+$, light blue). Similarly to HFB, vanillin also shows single-photon fragmentation events. The constants $c_1, \dots, c_4$ define the scale of the measured laser energy and are different for all four panels.

HFB clusters, the depletion beam splitting must result from single-photon fragmentation. Molecular dynamics simulations of these clusters using MMFF94 [20] show that a cluster will dissociate within a few picoseconds upon absorption of a single VUV photon and after the conversion of this energy into the vibrational degrees of freedom. A small cluster can even decompose in all its monomeric constituents. Photofragmentation in combi-

nation with ionization in the ToF-MS detector explains the absence of high cluster peaks with large n_{det} and the absence of clearly discernible Talbot orders in the normalized contrast [Fig. 2(a)]. In particular, charged fragments of larger clusters can account for the observed interference signal of the monomer in Fig. 2(c). Two-photon ionization of HFB in the gratings may also contribute to genuine monomer interference.

In contrast to HFB, the vanillin cluster data suggest a gradual transition from single-photon fragmentation to a mixture of single-photon ionization and fragmentation when the cluster number increases. This is consistent with the expectation that the cluster ionization energy decreases with the number of constituent molecules. Since fragmentation of vanillin clusters is less prevalent than for HFB, we can identify the first three Talbot orders in the mass spectrum of vanillin in Fig. 2(b). They are peaked around $n_{\text{det}} = 11$ ($m = 1672$ u), 5 (760 u), 2 (304 u) as determined by the pulse separation time. The maxima are shifted to higher masses with respect to the Talbot time because of the dipole force between the cluster polarizability and the laser light field. The fact that high- n vanillin clusters survive the ionization process supports the hypothesis that single-photon ionization competes favorably with photofragmentation at large n_{det} .

In conclusion, we have demonstrated a new class of matter-wave beam splitters which exploit the dissociation of composite objects for the coherent manipulation of particles. Here, fragmentation is triggered by the absorption of a single photon. Subsequent absorption events may occur but they modify neither the grating transmission function nor the diffraction pattern any further.

One might also invoke multiphoton ionization as an alternative to fragmentation for other classes of particles. Indeed, two-photon ionization at 266 - 280 nm can be a valid option for a range of aromatic molecules, including amino acids and polypeptides. However, single-photon processes are favorable to multiphoton schemes since they avoid nondepleting photoabsorption events and, therefore, maximize the interference contrast.

Compared to photoionization [8, 10] which can be applied to various types of atoms, clusters and molecules, fragmentation can be the dominant labeling process for weakly bound clusters, biomolecules or nanoparticles whose ionization energy exceeds the photon energy of the light grating. Photodepletion has already been successfully used for cluster spectroscopy, using visible [21, 22] or even infrared wavelengths [23–25].

One particularly well-suited example of particles susceptible to photofragmentation beam splitters are doped helium nanodroplets. Such nanodroplets, have been generated in the targeted mass range between 10^4 and 10^9 u [26, 27] and have been routinely used as nanocryostats for molecular spectroscopy [28]. At a typical temperature of about 380 mK the single-atom evap-

oration rate is low enough [29] not to induce any decoherence by particle emission during the 30 ms coherence time for OTIMA interferometry with 10^6 u. Moreover, at this temperature all vibrational modes of the dopant are essentially in their ground state and thermal decoherence is eliminated [30]. Photodepletion works exceedingly well in these systems [26] since the heat capacity of helium is low (7.2 K/atom) and the absorption even of a green photon suffices to evaporate more than 3000 helium atoms.

Optical fragmentation gratings may also open a new avenue to ion interferometry with composite particles. While mechanical diffraction structures have been successfully used for electron diffraction and interference [31], they may exhibit local patch potentials or charges. Optical masks can eliminate this problem, as successfully demonstrated with electrons [32]. Dissociation gratings are the most promising option for realizing absorptive gratings for highly charged composite systems.

Furthermore, photofragmentation gratings are interesting for many biomolecules. Most of them exhibit ionization energies in the range of 8-12 eV [33] and absorption would often rather induce fragmentation than ionization [34]. VUV induced dissociation is frequently used for mass spectroscopy [35, 36]. A similar mechanism may therefore also serve in realizing absorption gratings for biomolecules.

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A refined model for Talbot-Lau matter-wave optics with pulsed photo-depletion gratings

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We analyze time-domain Talbot-Lau interferometry of organic cluster beams that are exposed to pulsed photo-depletion gratings in the vacuum ultraviolet (VUV). We focus in particular on the analysis of the complex (phase and absorption) character of the optical elements. The discussion includes the role of wave front distortions due to mirror imperfections on the nanometer level and the effect of finite coherence in the diffraction gratings. This improved understanding of the interferometer allows us to extract new information on optical properties of anthracene and ferrocene clusters and to define conditions for future matter-wave experiments.

I. INTRODUCTION

Talbot-Lau interferometry has attracted great attention in the optics community since it allows one to coherently generate self-images of periodic structures using spatially incoherent light sources. This is of practical relevance in classical light optics [1, 2] and it opens new avenues in fields of research where coherent sources or refractive optical elements are not readily available. This holds for x-ray physics [3, 4], plasmonics [5] and ultrasound imaging [6]. For the same reason Talbot-Lau interferometry was proposed to be a promising tool for matter waves, in particular for very massive objects [7]. The idea has been successfully demonstrated in experiments with electrons [8], atoms [9–12] and hot molecules [13, 14].

Many of these earlier Talbot-Lau experiments were built using microfabricated diffraction gratings. However, complex particles of high polarizability are best manipulated using masks of light. This avoids the perturbing influence of van der Waals interactions or static charges that occur when particles transit material masks with slits as narrow as 100 nm [13, 15, 16]. In response to the need, standing light-wave phase gratings [17, 18], absorptive photoionization gratings [19] as well as photofragmentation gratings [20] have been introduced to quantum optics experiments with macromolecules and molecular clusters. Especially pulsed optical diffraction elements offer high versatility since the light-matter interaction can be timed with nanosecond precision and tuned via the laser wavelength, energy or pulse length.

Here we revisit the concept of optical time-domain matter-wave interferometry (OTIMA) which uses the periodic selection, diffraction and detection of nanoparticles in a series of three pulsed standing light-wave gratings [19, 21]. We focus on the practical limits imposed by the specifications of available laser systems and optical components at a wavelength of 157 nm. Particular attention is paid to the influence of a running wave that is superimposed to each standing light wave grating. This

offset is caused by the finite laser coherence and the mirror reflectivity. We further study the relevance of the local mirror roughness and demonstrate how to cope with global mirror surface deformations (flatness) on the level of 10 nm. The improved characterization of the OTIMA interferometer allows us to gain new information about optical cross sections and polarizabilities of particles such as molecular clusters of anthracene and ferrocene.

II. EXPERIMENTAL LAYOUT

The experimental sequence is as follows: Molecules are evaporated inside an Even-Lavie valve [22] in the presence of a high-pressure seed gas (1-7 bar of neon). A nozzle releases short (30 μ s) dense pulses of molecules that subsequently cool and cluster in the adiabatic expansion. With a mean velocity of around 900 ms^{-1} the particle beam is skimmed, collimated and transmitted into the interferometer chamber via two pumping stages. In our present work we focus on clusters of anthracene ($\text{Ac}_N = (\text{C}_{14}\text{H}_{10})_N$) and ferrocene ($\text{Fc}_N = (\text{C}_{10}\text{H}_{10}\text{Fe})_N$) since their volatilization and ionization properties are well suited for our analysis. For the measurements presented here, the molecular samples were heated to 430 K and 500 K, respectively. A typical mass distribution of ferrocene clusters is shown in figure 1b.

OTIMA interferometry, as depicted in figure 1a, combines three VUV photodepletion gratings (G_1 - G_3). These standing light wave gratings are formed by retro-reflecting three fluorine (F_2) excimer laser beams ($\lambda = 157.6 \text{ nm}$, $E \approx 3 \text{ mJ}$, $\tau = 8 \text{ ns}$) from the surface of a single 5 cm diameter CaF_2 dielectric mirror. Each beam has a flat top profile focused to an area of $A = 10 \times 1 \text{ mm}^2$ and neighboring beams are separated by 20 mm. The direction of the elongated waist is aligned with the cluster beam. At a photon energy of 7.9 eV (157 nm) the clusters efficiently ionize or fragment after absorption of a single photon. Since both processes deplete the particle beam, the gratings represent absorptive (beam depletion) masks with a period of $d = \lambda/2 = 78.8 \text{ nm}$. Clusters that survive the interferometer sequence are post-ionized with a fourth F_2 -laser pulse and detected by time-of-flight mass spectrometry. This allows us to compare particles with

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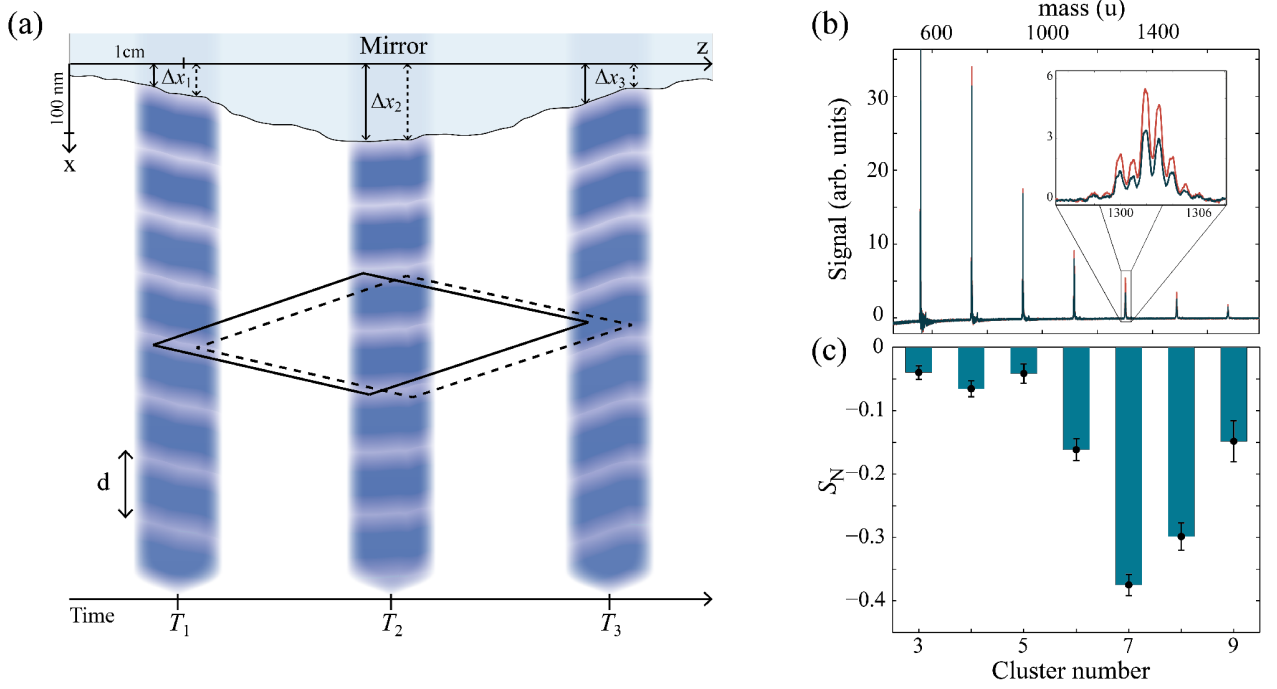


FIG. 1. Setup for cluster interferometry using three pulsed, absorptive standing light-wave gratings. (a) Mirror deformations shift the nodes of the standing waves within the laser spot. This reduces the fringe visibility due to the averaging over the phase-shifted interference patterns. This is indicated by the solid and dashed semiclassical paths for two particles that start with the same velocity and direction but at different positions. A mirror reflectivity $\mathcal{R} < 0.96$ and limited laser coherence are the reason why a running wave overlays the periodic gratings. The figure is drawn not to scale to illustrate the effects of minuscule mirror deformations. (b) Mass spectrum of ferrocene clusters that are detected after the third grating pulse using VUV photoionization in a time-of-flight mass spectrometer. We compare the interference signal S_{Int} (blue line) with a reference signal S_{Ref} (red line) as shown in the inset for the model system Fc_7 . (c) The normalized signal contrast S_N (see text) is shown for the clusters Fc_3 to Fc_9 .

different masses and optical properties in one and the same experimental run.

The absorptive character of grating G_1 confines the molecules in a comb of starting positions so that their wave functions expand over several nodes and antinodes of the standing light wave by the time the second grating is triggered. Rephasing of the wavelets behind G_2 leads to multipath interference and to the formation of a cluster density pattern, which can be resolved with the third grating pulse G_3 . This final pulse samples the interference pattern and transmits only those clusters whose wave function is aligned with the grating nodes. As in other realizations of Talbot-Lau interferometry the coherent self-imaging of the gratings occurs on a characteristic time scale, the Talbot time $T_T = md^2/h$, where m is the particle's mass, d the grating period and h Planck's constant.

We measure the fringe visibility by recording the transmitted number of particles behind G_3 as a function of the cluster mass. For this we choose a fixed pulse separation time T between two grating pulses and use two complementary measurement settings to visualize interference. In the interference setting the cluster signal

S_{Int} is recorded while the two pulse separation times $T = T_3 - T_2 = T_2 - T_1$ are kept equal to within $\Delta T < 1$ ns. Interference then enhances or reduces (depending on the phase) the cluster signal for masses close to the Talbot criterion. In the reference mode (S_{Ref}) we set an imbalance in the pulse separation times by $\Delta T > 50$ ns. For the given divergence and mass range in the molecular beam this is sufficient to wash out the interference pattern [23].

We define the cluster interference contrast as the normalized signal difference $S_N = (S_{Int} - S_{Ref})/S_{Ref}$, with $S_N \in [-1, 1]$ (Figure 1c). In the hypothetical case of a perfectly flat mirror surface and in the absence of external accelerations S_N is positive and equal to the theoretical visibility $\mathcal{V}$ of the interference pattern (see section IV). For a deformed mirror (see figure 1a), however, S_N is given by the visibility of the cluster density pattern at G_3 and the relative displacement $\Delta D = \Delta x_1 - 2\Delta x_2 + \Delta x_3$ of this Talbot image with respect to G_3 . Maximal constructive interference occurs for $\Delta D = zd$ and maximal destructive interference for $\Delta D = (z + 1/2)d$, with $z \in \mathbb{Z}$. The grating shifts Δx_i ($i = 1, 2, 3$) refer to a common zero line. In the present experiments the corrugated mirror

surface leads to negative S_N . Inertial or electromagnetic accelerations a may introduce additional fringe displacements that are, however, negligible for the parameters of our present experiments. If the mirror surface deformation exceeds 5-10 nm in the small region where the light pulse and the cluster cloud interact (i.e. 1-2 mm), different particles will experience differently shifted interferometers, as indicated by the two semiclassical paths in figure 1a. Averaging will thus lead to a reduced visibility of the density pattern at the position of G_3 . We account for the shift and reduction of the fringes by multiplying the theoretical visibility $\mathcal{V}$ (equation 7) by the fringe factor $\Omega \in [-1, 1]$, so that $S_N = \Omega \cdot \mathcal{V}$. On the one hand, this factor contains the interferogram shift for a single particle relative to G_3 . On the other hand, averaging many particles with slightly shifted interferograms is included, which leads to a reduction of the visibility at G_3 . Ω is determined by the mirror geometry in combination with the chosen pulse timings T_1, T_2, T_3 and the length of the detected cluster cloud.

III. MODELING FINITE MIRROR REFLECTIVITY AND FINITE LASER COHERENCE

The one-dimensional intensity profile of a standing light wave along x is ideally described by $I(x) = 4I_0 \cos^2(2\pi x/\lambda)$, with $I_0 = E/(A\tau)$ the incident laser intensity. In reality we need, however, to take into account imperfections of both the interferometer mirror and the grating lasers. At VUV wavelengths around 157 nm, state of the art CaF_2 mirrors are limited to a reflectivity of $\mathcal{R} = 96\%$. This may further degrade in the presence of hydrocarbon contaminations [24]. In addition, our free running F_2 lasers (EX 50, Gam Laser Inc., Orlando FL, USA) have a limited longitudinal and transverse coherence. They are expected to emit dominantly at 157.63 nm (ca. 90% of the total intensity) and 157.52 nm (ca. 10%) with a line width of $\Delta\lambda \approx 1$ pm [25]. This corresponds to a longitudinal coherence length of about 1 cm. The transverse coherence is determined by the width of the emitting aperture and amounts to 50-100 μm at the location of our interferometer mirror in 2 m distance from the laser exit.

These imperfections add an incoherent sum of running waves to the standing wave light field, which reduces the visibility V_λ of the laser grating. This reduces, in turn, the expected interference contrast of the diffracted matter-wave in dependence of the particle's VUV absorption cross section. Inhomogeneities within each grating, due to local roughness of the mirror surface, cause each individual particle to average over different standing wave phases during the finite laser pulse duration. Similarly, a divergent or tilted (with respect to the mirror surface) particle beam as well as mirror vibrations can lead to an averaging over grating phases during the 8 ns laser pulse duration. For example, particles with a ve-

locity of 1000 ms^{-1} orthogonal to the grating vector will accumulate over 8 μm of the standing light field within the laser pulse duration. Thus, a particle beam with a divergence of 1 mrad will average the field over 8 nm in the grating direction. These effects strongly depend on the absorption cross section of each particle at the laser wavelength λ .

To account for this running wave contribution in our model we distribute the incident laser intensity among a one-directional non-reflected contribution, $I(x) = (1 - \mathcal{R})I_0$, two counter-propagating running waves $I(x) = 2\mathcal{R}(1 - \mathcal{C})I_0$ and the standing light wave $4\mathcal{R}\mathcal{C}I_0 \cos^2(2x/\lambda)$. We quantify the coherent contribution by $\mathcal{C} \in [0, 1]$.

$$I(x) = (1 - \mathcal{R})I_0 + \mathcal{R} \left[2(1 - \mathcal{C})I_0 + 4\mathcal{C}I_0 \cos^2 \left(\frac{2\pi x}{\lambda} \right) \right] \quad (1)$$

Assuming Poissonian statistics the transmission function t is given by the cluster survival probability which is the probability of not absorbing any photon during the grating pulse. The mean number of absorbed photons per pulse depends on the optical absorption cross section σ_λ at $\lambda = 157$ nm. It is given by $n(x) = I(x)\lambda\sigma_\lambda\tau/(hc)$. We average the transmission function t over one grating period and get:

$$\begin{aligned} t &= \frac{2}{\lambda} \int_0^{\lambda/2} \exp[-n(x)] dx = \\ &= J_0 \left(i \frac{n_0}{2} \right) \exp \left[-n_0 \left(\frac{1}{4\mathcal{R}\mathcal{C}} + \frac{1}{4\mathcal{C}} \right) \right] \\ &\equiv J_0 \left(i \frac{n_0}{2} \right) \exp \left(-\frac{n_0}{2V_\lambda} \right) \end{aligned} \quad (2)$$

Here, J_0 denotes the zeroth order Bessel function and we introduce $n_0 = 4\mathcal{R}\mathcal{C}I_0\lambda\sigma_\lambda\tau/(hc)$ as the difference between the mean number of photons absorbed in the antinodes and the nodes of the standing wave. The visibility V_λ of the light grating is given by $V_\lambda = 2\mathcal{R}\mathcal{C}/(1 + \mathcal{R})$. If V_λ is known we can extract $n_0^{(i)}$ for every individual grating and all cluster masses separately by measuring the cluster transmission $t^{(i)}$.

IV. INFLUENCE OF THE SECOND GRATING

The established interferometer theory [26] describes the fringe visibility as a function of the molecular beam depletion in all three gratings and includes the dipole interaction in G_2 between the particle's optical polarizability α_λ and the electric laser light field. This allows us, in principle, to extract the particle's optical properties from the interference contrast as a function of the laser power in G_2 .

In a purely absorptive grating the difference between the mean number of absorbed photons in the antinodes and the nodes of the standing wave n_0 determines the effective open fraction of the grating and the interference

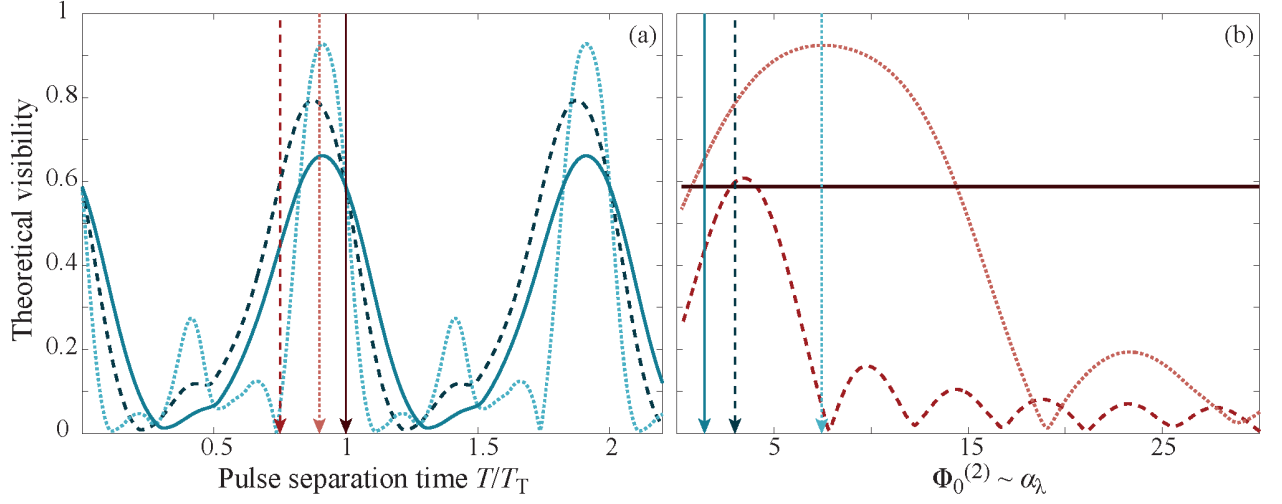


FIG. 2. Theoretical visibility $\mathcal{V}$ for different values of the optical polarizability. (a) Contrast as a function of the pulse separation time T , in multiples of the Talbot time T_T . It is plotted for varying phase shifts (polarizabilities) in the antinodes of the second standing wave ($\Phi_0^{(2)} = 7.5$, dotted; $\Phi_0^{(2)} = 3$, dashed; $\Phi_0^{(2)} = 1.5$, solid). (b) Contrast as a function of $\Phi_0^{(2)}$, i.e. for varying optical polarizability α_λ , plotted for three different pulse separation times ($T = 0.75 T_T$, dashed; $T = 0.9 T_T$, dotted, $T = T_T$, solid). For $T = T_T$ the particle polarizability does not affect the interference contrast at all while at $0.75 T_T$ the contrast oscillates with $\Phi_0^{(2)}$. The arrows and colors link plot (a) with plot (b). For both panels the transmission through all three gratings was fixed by setting $n_0^{(i)} = 6$.

contrast. The action of the first and the third grating in a Talbot-Lau interferometer is fully characterized by $n_0^{(1)}$ and $n_0^{(3)}$.

In the second grating, the electric field additionally imprints a relevant spatially dependent phase shift onto the traversing matter waves which is given by

$$\Phi(x) = \frac{4\pi^2 \alpha_\lambda \mathcal{R} C I_0 \tau}{hc} \cos^2 \left(\frac{2\pi x}{d} \right) \equiv \Phi_0^{(2)} \cos^2 \left(\frac{2\pi x}{d} \right) \quad (3)$$

where $\Phi_0^{(2)} = 4\pi^2 \alpha_\lambda \mathcal{R} C I_0 \tau / (hc)$ is the relative phase shift between two parts of the wave that pass the antinode and the node of G_2 , respectively.

It is convenient to introduce the dimensionless parameter β as the ratio of the absorption cross section and the optical polarizability α_λ

$$\beta = \frac{\lambda \sigma_\lambda}{8\pi^2 \alpha_\lambda} = \frac{n_0^{(2)}}{2\Phi_0^{(2)}}. \quad (4)$$

The signal behind the third grating in a symmetric Talbot-Lau configuration as shown in figure 1 can be Fourier expanded and expressed as a function of the relative grating shift ΔD , see [21] for details:

$$S(\Delta D) = \sum_{l=-\infty}^{\infty} B_{-l}^{(1)}(0) B_{2l}^{(2)} \left(l \frac{T}{T_T} \right) B_{-l}^{(3)}(0) \times \exp \left[\frac{2\pi i l}{d} (\Delta D - a T^2) \right] \quad (5)$$

where a denotes an external acceleration along the grating direction (negligible in the present experiments)

and $B_n^{(i)}$ are the Fourier coefficients that describe the influence of the individual gratings and are given by

$$B_n^{(i)}(\xi) = \exp \left(-n_0^{(i)} / 2 \right) \left(\frac{\sin \pi \xi - \beta \cos \pi \xi}{\sin \pi \xi + \beta \cos \pi \xi} \right)^{n/2} \times J_n \left[\operatorname{sgn} \left(\frac{1}{\beta} \sin \pi \xi + \cos \pi \xi \right) \times \frac{n_0^{(i)}}{2} \sqrt{\frac{1}{\beta^2} \sin^2 \pi \xi - \cos^2 \pi \xi} \right] \quad (6)$$

with the Bessel functions J_n .

The visibility of the fringe pattern is then defined as

$$\mathcal{V} = \frac{\max_x [S(x)] - \min_x [S(x)]}{\max_x [S(x)] + \min_x [S(x)]} \in [0, 1]. \quad (7)$$

If the pulse separation time fulfills the Talbot criterion $T = l \cdot T_T$ (with $l \in \mathbb{N}$) the interference pattern is unaffected by the phase contribution since all terms containing β in equation 6 drop out. In this case, the visibility $\mathcal{V}$ is only determined by $n_0^{(i)}$ of the three gratings as is seen in figure 2. For symmetric pulse delays deviating from the Talbot time, $T_2 - T_1 = T_3 - T_2 \neq l \cdot T_T$, the optical polarizability α_λ influences the interference contrast and β can be retrieved from a fit of the model to the data.

V. EXPERIMENTAL RESULTS

In order to extract the optical properties for all clusters several parameters need to be measured in the experi-

ment: An overall reduction of the interference contrast due to mirror deformations, common to all cluster numbers N , is included in the factor Ω . The visibility V_λ is assumed to be the same for all gratings since the laser beams are equally well aligned to the same mirror surface and the laser properties are known to be insensitive to the detailed laser conditions, such as its gas pressure or temperature [25]. The cluster interference contrast will, however, depend on V_λ since the grating quality influences the open fraction of the gratings $n_0^{(i)}$. Since the cluster size and structure are relevant, the parameter β must be fitted for every cluster number, individually.

In order to extract these parameters from our measurements, we proceed as follows: We record the normalized contrast S_N for all clusters in the accessible mass spectrum simultaneously as a function of the laser power in G_2 ¹. The laser pulse separation time T is set to the Talbot time of the hexamer ($T = 17.38 \mu\text{s}$ for ferrocene Fc_6 , $T = 16.65 \mu\text{s}$ for anthracene Ac_6) such that high contrast is expected around these masses (see figure 1). In addition, we record the transmission $t^{(i)}$ through every grating, individually, and feed this information into the theoretical model (Sections IV and III, see also [21]). The free parameters are adjusted until the theoretical predictions fit the experimental data. We start with one value of Ω and fit V_λ for the cluster that fulfills the Talbot criterion (the hexamer). This contrast depends only on the absorption in the standing wave ($n_0^{(i)}$), not on the optical polarizability α_λ as shown in figure 2. The resulting visibility V_λ and the measured grating transmissions are used to calculate $n_0^{(i)}$ with equation 2 for all cluster numbers N . With these values of $n_0^{(i)}$ the individual β is fitted for those cluster whose Talbot time does not match the chosen pulse separation time (here Fc_7 , Fc_8 , Fc_9 and Ac_7 , Ac_8 , Ac_9). The minimization of the least-square fit in V_λ and all β values is repeated for varying Ω to find the best match for all parameters. The results of this fitting procedure are shown in figure 3 for anthracene and figure 4 for ferrocene clusters. The routine converges very well and we find good agreement between the model and the experimental data.

Figure 3a displays the normalized contrast S_N for clusters between $N = 3$ -9, where the laser pulse energy in G_2 is varied to yield an average $n_0^{(2)}$ in the octamer Ac_8 of 3.5 (left panel), 2.3 (middle panel) and 1.6 (right panel). Even though the pulse delay time was set to match the Talbot time of Ac_6 we find the highest contrast at higher masses. This agrees with the expected shift of the resonance as a function of the particle polarizability and it in-

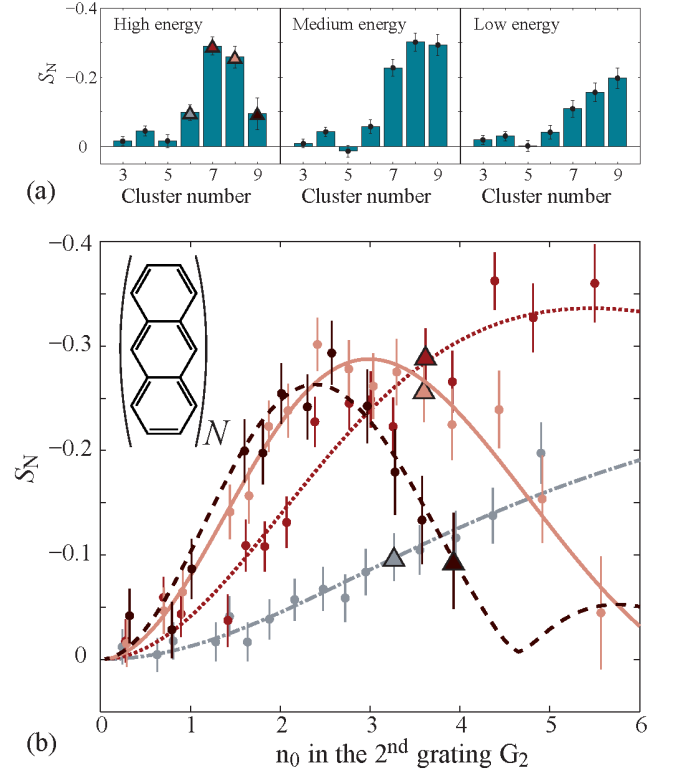


FIG. 3. Measured interference contrast as a function of the mean number $n_0^{(2)}$ of standing wave photons absorbed in the antinode of the second grating. (a) Interference measurements for three different laser energies reveal the shift of the maximal interference contrast towards higher masses for lower laser pulse energy. The triangles link the plots (a) and (b). (b) Normalized contrast for anthracene clusters of size $N=6$ -9: Ac_6 (grey, dashed-dotted), Ac_7 (red, dotted), Ac_8 (light red, solid), Ac_9 (brown, dashed). The lines are fits based on the model discussed in the text. Error bars represent one standard deviation of statistical error.

cludes the contribution of the running wave background. Figure 3b traces S_N as a function of the absorbed photon number in G_2 , for cluster composed of $N = 6, 7, 8, 9$ anthracene molecules.

The results of the model fit are collected in Table I. We find a value $V_\lambda = 0.75$ indicating high quality of the standing light waves at a distance of 1 mm to the mirror surface. Repeating the same experimental sequence with identical source settings and the mirror retracted to 2 mm to the cluster beam we find a decrease in the light grating visibility to $V_\lambda = 0.70$. This is consistent with a coherence length of the fluorine lasers of about 1 cm. The overall reduction of the fringe visibility by almost a factor of two compared to the ideal theory value shows that there is still room for development of high-quality VUV optics. The reduced contrast is partially due to the finite mirror reflectivity ($\mathcal{R} < 0.96$). An even larger influence is attributed to the finite mirror curvature. Independent measurements on different mirror species revealed a

¹ To attenuate the laser energy, we use the VUV absorption of oxygen [27] in a pressure cell that is filled with air. This cell is inserted into the evacuated beam guiding system, separated by two CaF_2 windows. It allows adjusting the transmitted laser power from a few percent up to 90% of the power at the cell entrance window, without compromising the timing performance or the beam profile.

Optical param.	Anthracene, mirror@1 mm	Anthracene, mirror@2 mm	Ferrocene, mirror@1 mm
V_λ	0.75 ± 0.02	0.70 ± 0.03	0.80 ± 0.01
Ω	-0.47 ± 0.03	-0.47 ± 0.04	-0.51 ± 0.01
β_7	0.53 ± 0.06	0.42 ± 0.05	0.56 ± 0.03
β_8	0.41 ± 0.03	0.39 ± 0.04	0.51 ± 0.02
β_9	0.40 ± 0.05	0.42 ± 0.05	0.41 ± 0.02

TABLE I. Optical parameters of the laser grating and the molecular clusters as extracted from a fit of the model to the data in Figure 3 and Figure 4. The errors refer to the statistical variations of the fits in Figure 3 and Figure 4. Substantially larger systematic variations may be caused by differences within the conformational structures of a given cluster mass. The first and the second data columns refer to the same set of experiments repeated for two different spatial separations between the laser mirror and the cluster beam.

global mirror curvature of about 6 km, bending the surface by up to 100 nm over 25 mm. Even this tiny displacement across the laser-cluster interaction zone (Figure 1) may cause measurable phase averaging.

To first order one might expect β to be independent of the cluster number N . For localized and non-interacting electrons both the absorption cross section and the polarizability should grow with the number of molecular constituents and their participating electrons. This assumption is essentially confirmed by Table I. One may see a small decrease in β from $N = 7$ to $N = 9$, both in the case of anthracene and ferrocene. But the data are also compatible with the observation that the β parameters vary by more than the one sigma systematic error of the values in table I when the experiment is repeated from day to day with changing source conditions. For similar source and interferometer settings the β parameters of anthracene were, however, found to be robust on the level of 10-20%, in all recent experiments.

[GIBT QUALITATIVE UEBEREINSTIMMUNG MIT MESSUNGEN VON 2012] *This finding seems to be in variance to data published in the first realization of an OTIMA interferometer, which was done in the same setup [19]. We attribute this difference to several factors: Marked differences in the nozzle setting, the source temperature and the conditions of the adiabatic expansion may change the average cluster conformation and cause the absorption cross section and polarizability to vary [28]. In addition, the earlier experiment and analysis [19] neglected the influence of finite laser coherence and finite mirror quality. The addition of the attenuation cell in the beam path of the G_2 laser allows us here to control the laser power in order to assess $n_0^{(2)}$ and the β parameter. There is good agreement between the model and many dozens of data points, as well as the reproducibility of the β parameters.*

Based on this successful concept we can also analyze the behavior of ferrocene clusters in OTIMA interferometry, as shown in figure 4. The data quality is comparable

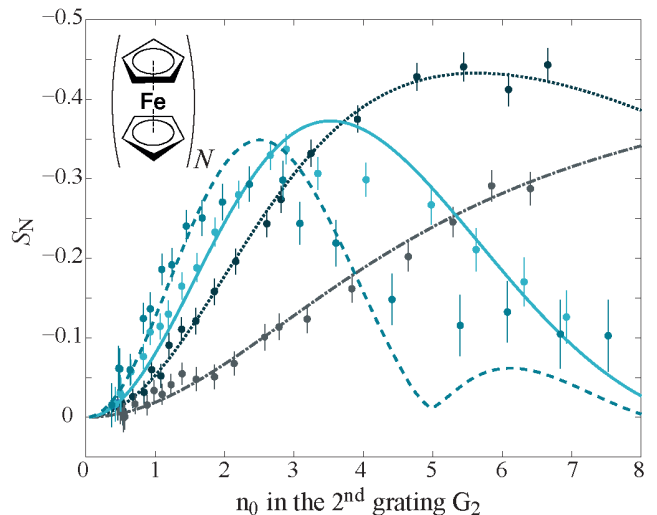


FIG. 4. Quantum interference of ferrocene clusters from $N=6-9$. It is shown as a function of number of absorbed photons in G_2 , which is also a measure for the open fraction of the grating. Fc6 (grey, dashed-dotted fit), Fc7 (dark blue, dotted fit), Fc8 (light blue, solid fit), Fc9 (blue, dashed fit). Error bars represent one standard deviation of statistical error.

to the case of anthracene, the model fits the data well and allows us to determine the optical cluster parameters. We find the light grating visibility V_λ to be consistent with the anthracene measurements [HIER NOCH SATZ ZU BESSERER VISIBILITY].

VI. CONCLUSION

We have presented a new analysis of matter-wave interferometry with pulsed photodepletion gratings in the vacuum ultraviolet regime. Our description takes into account real-world experimental limitations of the mirror flatness and roughness as well as the limited reflectivity of the VUV optics. This is essential for a better understanding of the OTIMA concept, which is a generic idea for matter-wave interferometry with complex nanoparticles. Our method allows us to extract for the first time quantitatively the β parameter of organic clusters, i.e. the ratio of their absorption cross section and polarizability at 157 nm. This is remarkable as it is generally demanding to assess optical properties of weakly bound van der Waals clusters in the vacuum ultraviolet.

Matter-wave experiments have been proposed to test the linearity of quantum mechanics with very massive particles [26, 29–32]. Such studies require the best possible understanding of all mechanisms that may cause an experimental deviation from theoretical predictions. This includes quantum decoherence in the presence of molecular collisions, scattering or emission of photons [14]. The good quantitative description that could be achieved here, underlines that it is promising to fur-

ther proceed with explorations of high-mass quantum interference.

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