Deliverable 4.5:
Optical depletion gratings

Objectives and description of Tasks

Report on pulsed optical depletion gratings for complex molecules/nanoparticles.

Work performed and achievements

Our efforts in developing the best suited nanomechanical mask (see D2.1 and D4.1) for quantum diffraction experiments with nanoparticles have shown that particle-wall interactions may impose limits on high-mass quantum interference experiments. It is therefore important to establish methods that can serve the purpose of absorptive masks without introducing van der Waals forces.

Pulsed optical depletion gratings turn out to fill this gap in an ideal way since standing light waves have a high reproducibility and periodicity. Their appearance time and opening fraction can be easily controlled. In addition, pulsed experiments allow the elimination of many dispersive phase shifts that would limit high mass quantum interference.

We have studied photo-depletion gratings experimentally and theoretically at UNIVIE in close collaboration with UDE and UNIBAS (see also D4.2). Different depletion mechanics based on absorption of either a single photon or two photons in the antinodes of a standing wave have been explored and shall be presented here.

Single photon gratings in the OTIMA interferometer

Our most recent realization of a near-field matter-wave experiment for nanoparticles at UNIVIE operates in the time domain and is called OTIMA (Optical TIme-domain MAtter-Wave). It is based on a sequence of three UV light gratings that prepare the necessary coherence in the molecular beam, induce diffraction and sample the interferogram. We use three pulsed fluorine lasers that emit light at a wavelength of 157 nm and reflect it off a single mirror so that every beam forms a standing light wave gratings with a periodicity of 78.5 nm [1]. All laser beams are rigidly phase coupled through the common mirror.

Photo-depletion gratings are measurement-induced gratings[2] as they determine a comb of positions where the particles can pass. Here we are particularly interested in ‘universal’ gratings in the sense that they are not sensitive to the detailed electronic structure of the diffracted particles. Depending on the choice of the nanoparticle species, the possibility of absorption of a single or many photons in the antinodes of the standing wave may create an absorptive mask: Nanoparticles can be for instance be removed from the molecular beam by ionization, if the ionization energy is well below the energy of a single photon, here 7.9 eV (see Fig. 1a). Even more generally optically induced fragmentation of weakly bound clusters or photocleavage of tailored compounds may lead to a depletion of the delocalized wave function at the antinodes of the standing light wave.

Single photon ionization gratings

Recently, we have successfully employed *single-photon ionization gratings* in the OTIMA experiment which has led to the first demonstration of quantum interference of *clusters of organic molecules*, as shown in Figure 1b. High contrast interference experiments with clusters of anthracene with de Broglie wavelengths as tiny as 200 fm reveal the power of this concept which shall be extrapolated to higher mass particles in the coming funding period.

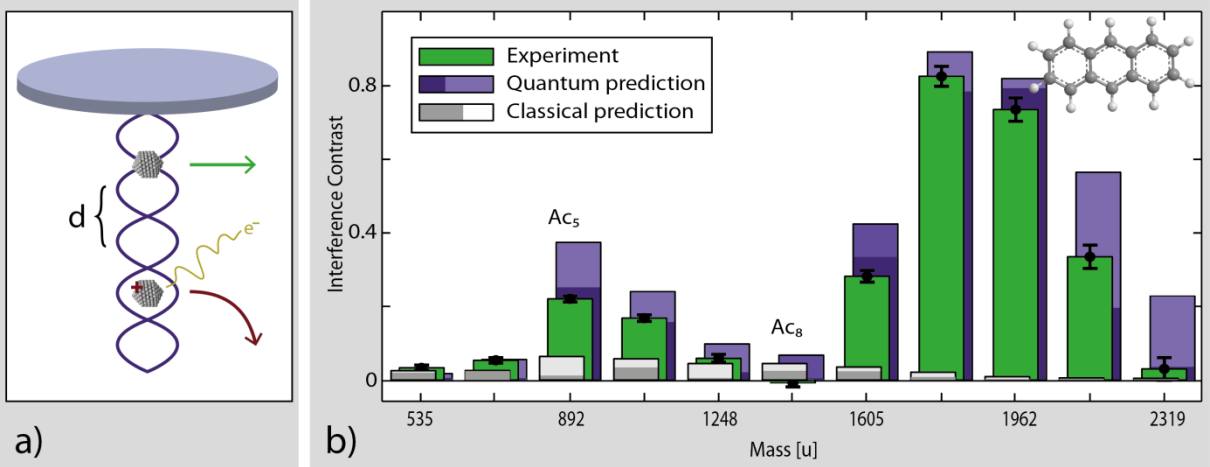

Figure 1: *a) Single photon ionization in the antinodes of a standing light wave removes particles selectively since the created ions are deflected out of the molecular beam by electric fields.*
b) For clusters of anthracene molecules quantum interference has been successfully demonstrated for the first time in the OTIMA setup[1]. The bars correspond to different clusters of this aromatic molecule, ranging here from Ac_3 to Ac_{13}. For a fixed pulse delay between the grating lasers we observe high contrast for clusters whose mass match the chosen timing. The experimental data matches the quantum theoretical predictions but strongly deviates from the contrast that could be explained by classical shadow effects[3].

Single-photon ionization is expected to be applicable to a wide class of nanoparticles. Possible candidates are metal clusters that we can produce in UNIVIE or functionalized organic molecules and silicon spheres synthesized at UNIBAS.

An ionizing optical diffraction grating does not only act as an absorptive mask but also always imprints a spatially periodic phase on the molecular wave due to the interaction between the electric laser light field and the particle's optical polarizability. The expected interference fringe pattern therefore depends not only on the photoionization cross section σ_{ion} but also on the polarizability α at λ=157 nm. We combine these two properties in the parameter: $\beta = \sigma_{ion}\lambda / 8\pi^2\alpha$, that determines the quantum visibility.

The OTIMA interferometer is built from three equal laser pulses with a well-defined pulse separation time. The first grating prepares the cluster coherence, the second grating is responsible for matter wave diffraction and the third pulsed standing light wave probe the cluster density pattern that forms because of quantum interference. The fringe pattern forms, however, resonantly, only in a time window, which is in practice limited to a width of several dozen nanoseconds.

Quantum interference reveals itself in four different ways (see [1, 4]), first by a modulation of the transmitted mass spectrum, which also encodes the Talbot resonance of near-field matter wave interferometry. For particles of similar velocity, mass encodes the de Broglie wavelength, also. We see in Fig. 1b an enhancement of the cluster transmission at masses around 900 amu and 1800 amu which is compatible with the expected Talbot time and Talbot length.

Interference reveals itself also in a spatial modulation of the cluster distribution, in a resonant appearance of the and in the detailed dependence on the symmetry of the pulse sequence. All these phenomena have been found in the experiment – which demonstrates that single-photon absorption/ionization gratings are perfect depletion gratings for complex interferometric tasks.

However, for a full understanding of the observed interference signals, imperfections of the optics and the standing waves have to be taken into account. Present-day technology neither allows the fabrication of VUV mirrors with a reflectivity of better than 96 % at 157 nm nor perfect flatness to better than 5 nm over the entire area mirror. This would be necessary to ensure the same boundary condition for all three standing light waves.

Phase shifts therefore have to be taken into account when the grating beams (pulse times) are moved across the mirror. Also the running wave background that is caused both by the limited mirror reflectivity and the limited coherence of molecular fluorine lasers[1] has to be included in the modelling of the quantum fringes. This running wave component superposes the standing light wave and contributes differently for every cluster to the measured grating transmission.

A thorough understanding of this influence on the achievable contrast as a function of the cluster number has been achieved over the last months. It now allows us – by measuring interference and transmission curves – to extract not only the individual β-parameters of all anthracene cluster, but also to measure the quality of the standing wave and the phase of the interference signal.

Single-photon fragmentation/dissociation gratings

In the same setup, we have also performed interference experiments with clusters of hexafluorobenzene (HFB, see D2.5) which has an ionization energy of about 10 eV – which is well above the energy of a single VUV photon in the light gratings. Still, we can realize absorptive masks for clusters of this molecule since HFB has a strong absorption band around 157 nm and weakly bound van der Waals clusters easily fragment upon photon absorption.

In Fig. 2c we show preliminary quantum interference results for small clusters and we prove that photofragmentation after absorption of a single photon is dominates the grating character.

We do this by first studying the signal as a function of the *detection* laser power. This laser is of the same type as the grating lasers (157 nm fluorine laser) and it is used to post-ionize the particles behind the interferometer before they are extracted into a time-of-flight mass spectrometer.

[1] The VUV F2 lasers have a longitudinal coherence of ~ 1 cm and a transverse coherence of some 10 µm.

We show that indeed two photons are needed to ionize the molecular clusters by observing a curve with positive curvature in the low power regime. This is a typical sign for a multi-photon absorption process.

We then study the transmission through a single grating as a function of the *grating* laser power and detect a typical single photon absorption behavior which can now only be explained by fragmentation.

These datasets are shown in Figure 2a for the 4-fold cluster of hexafluorobenzene. In addition, we can provoke observable photofragmentation with the detection laser by increasing its photon density in the ionization region.

Fragmentation of the clusters manifests itself in a decrease of signal with increasing laser power as shown in Figure 2b.

Non-ionizing depletion gratings bear the potential of being particularly well suited for particles of very high ionization energy but also for high mass experiments. The possibility of avoiding the technologically difficult VUV wavelength range at 157 nm – where coherent intense laser beams and high-quality optics are demanding – is an attractive motivation for exploring depletion gratings at $\lambda \geq 200$ nm in the next funding period, too.

Conclusions

Photodepletion gratings have been implemented experimentally as ionization gratings for Anthracene clusters as well as in the form of cluster fragmentation / molecular emission gratings in the OTIMA interferometer at UNIVIE.

The deliverable can therefore be counted as successfully accomplished.

Outlook

Future work will build on this success and explore photodepletion in a more general context in far-field experiments as well as near-field interferometry. In particular photo-cleavage gratings are of interest for future experiments also on otherwise challenging biomolecular samples.

Reference

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[4] S. Nimmrichter, P. Haslinger, K. Hornberger, M. Arndt, Concept of an ionizing time-domain matter-wave interferometer, New J. Phys., 13 (2011) 075002.

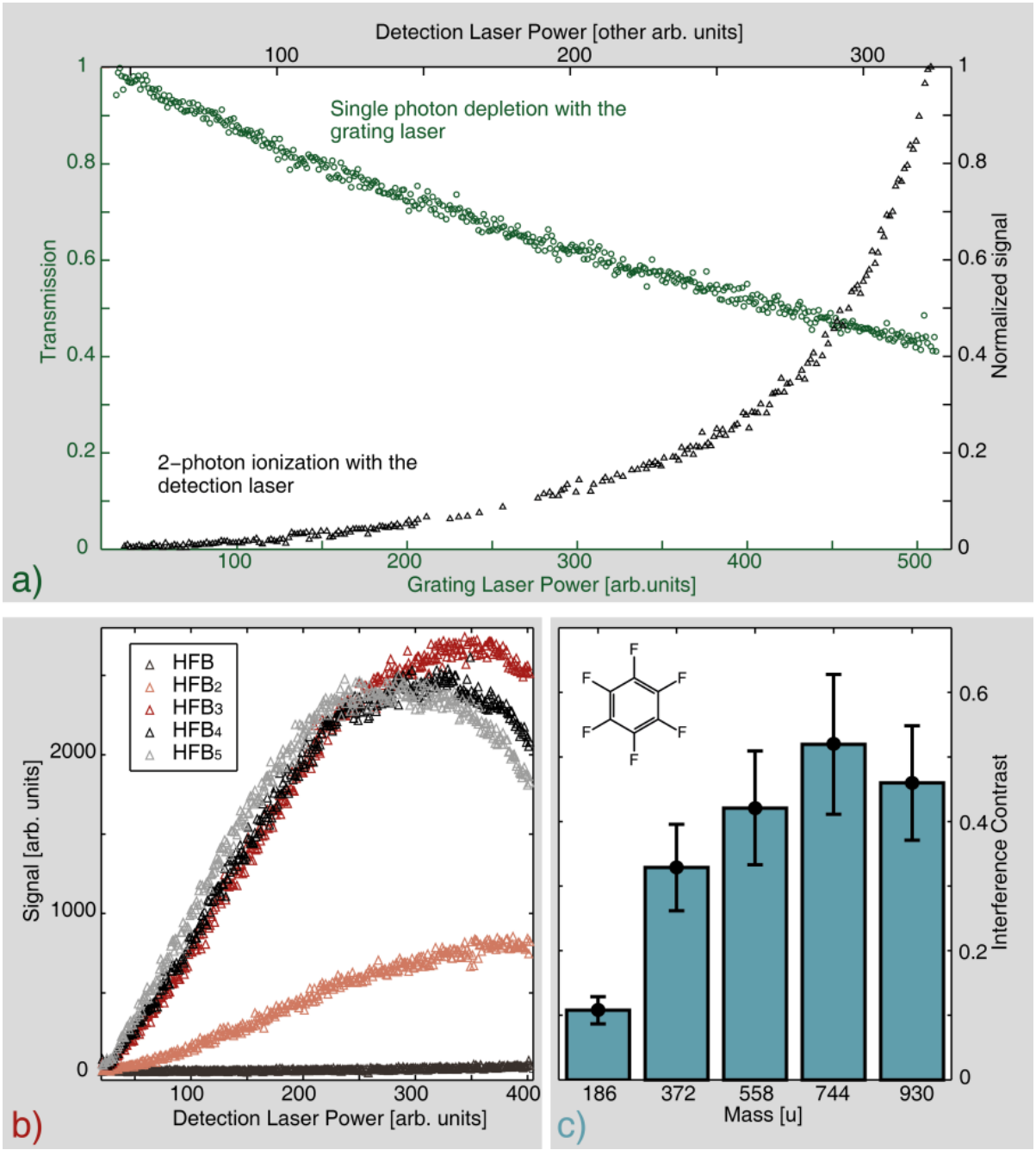

Figure 2: *Preliminary results on photofragmentation gratings with clusters of hexafluorobenzene. a) Data for the 4-fold cluster. It shows a single photon absorption behavior in the transmission through the grating (green circles), but a two-photon ionization curve at the detection (black triangles) – in the usually employed low power regime of the detection laser. b) Detection signal as a function of the detection laser power in the high power regime for the single HFB molecule to the 5-fold cluster. The larger the cluster the earlier the signal saturates and the cluster fragments for increasing laser power. C) Quantum interference contrast of clusters of hexafluorobenzene. The unexpected contrast for the monomer (at 186 amu) is an artifact of fragments produced in the gratings that reach the detector.*