Diffraction of Complex Molecules by Structures Made of Light

Olaf Nairz, Björn Brezger, Markus Arndt, and Anton Zeilinger

Universität Wien, Institut für Experimentalphysik, Boltzmanngasse 5, A-1090 Wien, Austria
(Received 1 June 2001; published 26 September 2001)

We demonstrate that structures made of light can be used to coherently control the motion of complex molecules. In particular, we show diffraction of the fullerenes C_{60} and C_{70} at a thin grating based on a standing light wave. We prove experimentally that the principles of this effect, well known from atom optics, can be successfully extended to massive and large molecules which are internally in a thermodynamic mixed state and which do not exhibit narrow optical resonances. Our results will be important for the observation of quantum interference with even larger and more complex objects.

The great success of atom optics has stimulated the question whether it is possible to extend the methods developed in this field to more complex and massive quantum objects. In this Letter we demonstrate for the first time the coherent control of the molecular motion using optical structures for macromolecules.

One possible manipulation technique for molecules has been demonstrated earlier with the use of material nanostructures. They have, for example, successfully been used in atom interferometry [1], in the determination of the bond length of a helium dimer [2], and more recently in interference experiments with the fullerenes C_{60} and C_{70} [3,4]. The use of solid nanostructures has the advantage of being universal and largely independent of the detailed internal character of the diffracted object. However, since the structure dimensions have to be of the order of 100 nm, material devices are extremely fragile and can be blocked or destroyed by the molecules.

In contrast to that, diffraction structures made of light are promising alternatives: The periodicity can be perfect, the transmission is high, and one can realize different types of gratings. Light may act as a real or imaginary index of refraction for matter waves and thus form a phase grating or an absorption grating.

For atoms, amplitude gratings can be based on various effects which lead to an effective spatially periodic depletion of relevant states of the atomic beam [5,6]. For example, the extraction of particles using ionization in a standing wave is conceivable.

Phase gratings based on the nondissipative dipole force were demonstrated for the diffraction of atomic beams both in the thin grating or Raman-Nath regime [7] and in the thick grating or Bragg regime [8]. They have been successfully implemented to build up complete Mach-Zehnder interferometers [9,10] and they find applications in atom lithography [11,12] or the manipulation of Bose-Einstein condensates [13]. Combinations of absorptive and phase structures can also be used for complex blazed gratings [14].

In spite of the great success in atom optics, light gratings have not yet been applied to larger molecules. This is mainly due to significant differences between atoms and

molecules which have to be taken into account in a practical realization: The optical linewidths of large molecules are typically of the order of 100 THz instead of a few MHz in the atomic regime. In the case of fullerenes the electrical polarizability, mediating the coupling to the optical field, remains constant within a factor of 2 throughout the whole frequency range from dc to UV. The high complexity of these molecules leads to non-negligible absorption from the ultraviolet well into the visible wavelength region and for the realization of a pure phase grating one would think that absorption should be excluded. This reasoning is based on the experience from atom optics that absorption is usually followed by spontaneous emission, which carries which-path information into the environment.

In the following we will demonstrate that the principles of light gratings can actually be successfully carried over to fullerenes, which are internally in a thermodynamic mixed state. Even in the presence of absorption the fullerenes may maintain coherence between states of equal energy since the absorbed quanta are trapped in the molecules.

A schematic of the setup is shown in Fig. 1. Essential parts of the setup are already described in [3] and an in-depth characterization of the detector is given in [4]. A ceramic oven containing the fullerene powder is heated
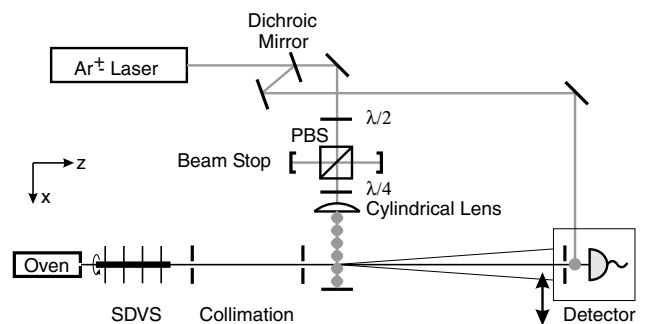

FIG. 1.   An oven generates a fullerene beam. Collimation and velocity selection prepare the required transverse and longitudinal coherence. The beam interacts with a circularly polarized standing light wave of periodicity 257 nm. The power is variable between 0 and 9.5 W and focused to $w_z \times w_y = 0.05 \times 1.3$ mm^2. The molecular diffraction pattern after the light grating is recorded by a scanning optical ionization stage.

to approximately 900 K. The fullerene beam exits with a most probable velocity of 200 and 170 m/s in the case of C_{60} and C_{70}, respectively, both with a FWHM velocity spread of $\triangle v/v \approx 0.6$. In order to separate the individual diffraction peaks in our experiment we select only a slow part of the velocity distribution using a slotted disk velocity selector (SDVS) [15]. We select a most probable velocity of approximately 120 m/s with a velocity spread of $\triangle v/v \approx 0.17$, which allows us to keep about 6% of the initial molecular flux. The beam is collimated by two vertical slits of 7 and 5 μm separated by 1.13 m.

Both the standing light wave and the detection are implemented using the same 27 W, multiline visible Ar^+ laser. Using a dichroic mirror with a transmission of $T > 76\%$ at $\lambda_L = 514.5$ nm and $T < 1\%$ for all other laser lines we isolate the single green line for the standing wave from the color mixture used in the ionization. The green power can be varied continuously up to 9.5 W by rotating a half-wave plate in front of a polarizing beam splitter (PBS). The quarter-wave plate between the polarizing beam splitter and the mirror is set to 45° so that the back-reflected light is deflected by the PBS and cannot return into the laser. The ensemble averaged molecule-light interaction is expected to be insensitive to the light polarization since the molecules enter in a thermal mixture of rotational and vibrational states.

In order to ensure that the light potential seen by the fullerenes does not average out during their transit through the standing light wave, the nodal planes of the standing light wave have to be parallel to the molecular beam. This is facilitated by focusing the laser beam in the direction of the molecular trajectory. The cylindrical lens has a focal length of 30 cm and reduces the laser beam from a $1/e^2$ radius of 1.3 mm to $w_z \approx 50$ μm at the mirror surface. The final alignment of the mirror could be achieved with the collimated fullerene beam itself. The mirror was first moved into the fullerene beam, so that it cut a part of it, and was then rotated around the axis centered on the mirror surface and normal to both the fullerene beam and the laser beam. A symmetric decrease in count rate when rotating the mirror in both directions indicated the correct alignment. After this procedure we estimate the mirror to be parallel to the molecular beam by better than 0.5 mrad. During the experiment the beam passed the mirror surface in a distance of 100 μm.

The height of the detected fullerene beam was determined using a knife edge and amounted to (625 ± 70) μm FWHM. A coarse overlap between the molecular beam and the standing wave was achieved using a horizontal slit. The final alignment was done using the diffraction pattern itself: maximal diffraction efficiency indicated perfect overlap.

In order to record the diffraction pattern 1.2 m downstream we scanned the focused detection laser beam (power 17 W, beam waist ≈ 4 μm) across the molecular beam in 2 μm steps. The fullerenes were ionized by absorption of 20 to 30 blue and green photons and the ions were subsequently counted. The width of the detector at this power for C_{60} is approximately 6 μm [4]. For C_{70} the absorption cross section is much higher than for C_{60} and so the width of the detector would be larger. In order to ensure a comparable detector width for both types of fullerenes, we placed a 5 μm precision slit in front of the laser beam and moved it together with the focusing optics.

Since the whole vacuum apparatus drifted by several micrometers within a few hours we had to limit the recording time per diffraction pattern to less than an hour. In order to obtain a sufficient counting statistics we performed several (typically 15) runs. Within each run we recorded both an undiffracted and a diffracted picture by blocking and unblocking the standing light wave for each individual detector position. The reference pictures allowed us to center the diffraction patterns before they were summed. The far field diffraction patterns were recorded at different laser powers both for C_{60} and C_{70} as shown as full squares in Figs. 2 and 3.

The continuous curves in these graphs are the result of our theoretical model: The fullerenes do not have a permanent electric dipole moment but a finite polarizability α.
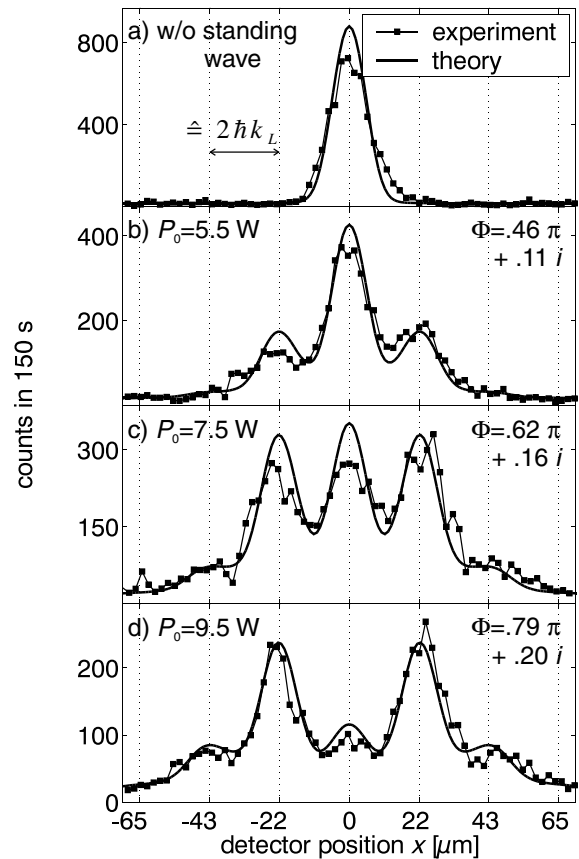

FIG. 2. Interference patterns for C_{60} for different laser powers. Φ is the phase shift parameter as defined in Eq. (3). Twice its imaginary part gives the mean absorbed photon number [Eq. (5)]. The diffraction efficiency into each of the first diffraction orders in case (d) can be estimated to be about 25%.
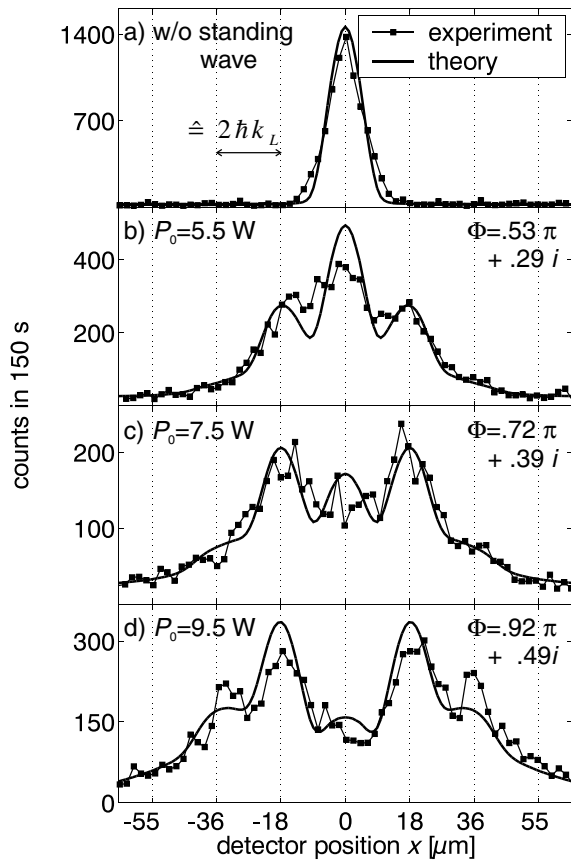
FIG. 3. Interference patterns for C_{70} for the same laser powers as in Fig. 2.

The electric field E of the laser induces in first order a dipole moment and in second order a potential $V^{\mathrm{dip}}(x, z) = -\frac{1}{2}\alpha\overline{E^2(x, z, t)} = -\alpha I(x, z)/2c\varepsilon_0$, proportional to the local intensity $I(x, z)$.

The action of the standing light field on the molecular de Broglie waves is described as that of a thin grating in the Raman-Nath regime [7]: the displacement of the molecules due to diffraction and within the light field itself is much smaller than one grating period. Immediately behind the grating the spatial distribution of the molecules is therefore essentially unchanged. We thus obtain the following transmission function [16] for those molecules that leave the interaction zone in the same internal state as before:

$$t^{\mathrm{dip}}(x) = \exp\left(-\frac{i}{\hbar}\int V(x, z(t))\, dt\right) \qquad (1)$$

$$= \exp[2i\Phi\cos^2(k_L x)], \qquad (2)$$

where the phase shift has a mean value of

$$\Phi = \sqrt{\frac{2}{\pi}}\frac{P_0\alpha}{w_y v_z \hbar c\varepsilon_0}. \qquad (3)$$

Equation (3) holds for the passage of the molecules through the center of an elliptical Gaussian laser profile of vertical $1/e^2$ width w_y as in the experiment. P_0 is the power of each constituent running wave and v_z the mole-

cule velocity. By virtue of the grating periodicity in x with $\lambda_L/2$, far field peaks deflected by $\Delta p_x = \pm m 2\hbar k_L$ ($m \in \mathbb{N}$) result. Their relative intensities are calculated by the Fourier decomposition of Eq. (2) and are given by the squared Bessel functions $J_m^2(\Phi)$. In particular, the zeroth diffraction order can be suppressed for $J_0(\Phi) = 0$—in contrast to diffraction at a mechanical grating. The experimental parameters for Fig. 2d are close to this situation.

So far we have neglected the imaginary part of the polarizability α which describes the absorption of grating photons by the molecules with a cross section $\sigma = \mathrm{Im}(\alpha)k_L/\varepsilon_0$. Absorption processes change both the internal state and the momentum of the molecule but lead neither to a loss of signal nor to spontaneous emissions.

Those molecules that absorb one or more photons will change to a different internal state but still contribute to the detected signal. We take this into account by introducing transmission functions $t_{0\to n}(x) = t^{\mathrm{dip}}(x) \cdot t_{0\to n}^{\mathrm{abs}}(x)$ and an incoherent sum over the contributions for different absorbed photon numbers n. The dipole part $t^{\mathrm{dip}}(x)$ is taken to be the same for all n, where we replace α with $\mathrm{Re}(\alpha)$ in Eq. (3). The absorptive part follows from assuming a Poissonian statistics $p_{\bar{n}(x)}(n)$ with a mean absorbed photon number $\bar{n}(x)$:

$$\bar{n}(x) = \frac{\sigma}{\hbar\omega_L}\int I(x, z(t))\, dt \qquad (4)$$

$$= \mathrm{Im}(\Phi)4\cos^2(k_L x) \qquad (5)$$

$$t_{0\to n}^{\mathrm{abs}}(x) = \sqrt{p_{\bar{n}(x)}(n)}\left(\frac{E(x, z)}{|E(x, z)|}\right)^n. \qquad (6)$$

The use of Poissonian statistics is equivalent with assuming that the absorption processes are independent. This is justified by the—compared to the inverse absorption rate—fast internal relaxation processes. The first factor in Eq. (6) conserves the number of molecules since $\sum_{n=0}^{\infty}|t_{0\to n}^{\mathrm{abs}}(x)|^2 = 1$. The second factor, representing a phase independent of z, stems from momentum conservation: The molecules inherit the phase of the absorbed photons. In effect, the transmission function $t_{0\to n}(x)$ contains only Fourier components corresponding to odd (even) multiples of $\hbar k_L$ for odd (even) n. In our experiment the resolution is good enough to clearly resolve a peak spacing of $2\hbar k_L$. Peaks at odd multiples of $\hbar k_L$ fill in the minima and decrease the contrast of the interference pattern.

The simulation is based on the described transmission functions followed by free-space propagation and it contains no free parameters. An incoherent sum is performed over source points in the first slit, over the measured longitudinal velocity distribution, and over the measured vertical fullerene distribution in the plane of the grating. We take into account the vertical laser profile and the finite detector resolution. The presented formalism relies on the assumption that all populated molecular states have the same complex polarizability and that all internal states are detected with the same efficiency. The complex ground

state polarizabilities at 514 nm which enter our model are $\alpha_{C60} = (101 + 8i)$ Å$^3 \times 4\pi\varepsilon_0$ [17] and $\alpha_{C70} = (118 + 20i)$ Å$^3 \times 4\pi\varepsilon_0$ [18]. The imaginary parts correspond to absorption cross sections of $\sigma_{C60} = 1.2 \times 10^{-17}$ cm^2 and $\sigma_{C70} = 3.1 \times 10^{-17}$ cm^2.

Both Figs. 2 and 3 show a very good overall agreement between the theoretical model and our experimental results. Some broadening in the experimental curves is attributed to nonperfect angular alignment of collimation and detection slits and similar imperfections. Only in the case of C_{70} interacting with the maximum laser power (Fig. 3d) we see a surprising deviation between the experiment and our model. The diffracted intensity at $\pm 4\hbar k_L$ seems to be higher in the experiment than in the model. The significant difference of C_{70} as compared to C_{60} is its higher absorption cross section for the green light—therefore in the simulation at maximum laser power 12% of the C_{70} molecules absorb two photons, thus also contributing to the peaks at even multiples of $\hbar k_L$, but only 4% in the case of C_{60}. The effect could even be stronger due to the higher absorption cross section [19] or a higher polarizability of the excited states.

Concluding, we have shown that optical gratings for fullerenes work very well using intense visible laser light. The diffraction patterns can be fully understood when the standard theory for dipole forces is supplemented by the effect of photon absorption. Optical gratings are generally important for quantum interference experiments with even larger and more complex objects—under the general requirement of not too prominent absorption at the employed laser wavelength—since they have an excellent regularity, transmission, and efficiency and cannot be destroyed by the diffracted particles. In particular, we have shown that a C_{60} beam can be split at will in two or three arms with good efficiency (cf. Figs. 2c and 2d). This is important in future applications of interferometers using beam splitters based on light.

Optical gratings possess useful scaling properties: Mass and polarizability have roughly the same scaling behavior. They are both proportional to the volume of the object. Conceptually, light gratings may ultimately even be used for particles the size of which is comparable to the grating constant. It will also be interesting to see the influence of, for instance, the symmetry of the molecules or their internal excitation on their ability to interfere. Finally, one may envisage molecule holography with dedicated light structures.

[1] D. W. Keith, C. R. Ekstrom, Q. A. Turchette, and D. E. Pritchard, Phys. Rev. Lett. **66**, 2693 (1991).
[2] R. E. Grisenti, W. Schöllkopf, J. P. Toennies, G. C. Hegerfeldt, T. Köhler, and M. Stoll, Phys. Rev. Lett. **85**, 2284 (2000).
[3] M. Arndt, O. Nairz, J. Voss-Andreae, C. Keller, G. van der Zouw, and A. Zeilinger, Nature (London) **401**, 680 (1999).
[4] O. Nairz, M. Arndt, and A. Zeilinger, J. Mod. Opt. **47**, 2811 (2000).
[5] R. Abfalterer, C. Keller, S. Bernet, M. K. Oberthaler, J. Schmiedmayer, and A. Zeilinger, Phys. Rev. A **56**, R4365 (1997).
[6] S. Kunze, K. Dieckmann, and G. Rempe, Phys. Rev. Lett. **78**, 2038 (1997).
[7] P. E. Moskowitz, P. L. Gould, S. R. Atlas, and D. E. Pritchard, Phys. Rev. Lett. **51**, 370 (1983).
[8] P. J. Martin, B. G. Oldaker, A. H. Miklich, and D. E. Pritchard, Phys. Rev. Lett. **60**, 515 (1988).
[9] E. M. Rasel, M. K. Oberthaler, H. Batelaan, J. Schmiedmayer, and A. Zeilinger, Phys. Rev. Lett. **75**, 2633 (1995).
[10] D. M. Giltner, R. W. McGowan, and S. A. Lee, Phys. Rev. Lett. **75**, 2638 (1995).
[11] G. Timp, R. Behringer, D. M. Tennant, J. E. Cunningham, M. Prentiss, and K. K. Berggren, Phys. Rev. Lett. **69**, 1636 (1992).
[12] Th. Schulze, T. Müther, D. Jürgens, B. Brezger, M. K. Oberthaler, T. Pfau, and J. Mlynek, Appl. Phys. Lett. **78**, 1781 (2001).
[13] L. Deng, E. W. Hagley, J. Denschlag, J. E. Simsarian, M. Edwards, C. W. Clark, K. Helmerson, S. L. Rolston, and W. D. Phillips, Phys. Rev. Lett. **83**, 5407 (1999).
[14] C. Keller, M. K. Oberthaler, R. Abfalterer, S. Bernet, J. Schmiedmayer, and A. Zeilinger, Phys. Rev. Lett. **79**, 3327 (1997).
[15] *Atomic and Molecular Beam Methods,* edited by G. Scoles (Oxford Univ. Press, Oxford, 1988).
[16] C. S. Adams, M. Sigel, and J. Mlynek, Phys. Rep. **240**, 143 (1994).
[17] Via the Clausius-Mosotti relation [18] from Fig. 2 in E. Sohmen, J. Fink, and W. Krätschmer, Z. Phys. B **86**, 87 (1992).
[18] From Table 2 in J. U. Andersen and E. Bonderup, Eur. Phys. J. D **11**, 435 (2000).
[19] H. Gratz and A. Penzkofer, Photochem. Photobiol. **127**, 21 (1999).