Experimental verification of the Heisenberg uncertainty principle for fullerene molecules

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The Heisenberg uncertainty principle for material objects is an essential corner stone of quantum mechanics and clearly visualizes the wave nature of matter. Here, we report a demonstration of the Heisenberg uncertainty principle for the fullerene molecule C_{70} at a temperature of 900 K. We do this by showing the increase in molecular momentum spread after passage through a narrow slit with a variable width down to 70 nm. We find good quantitative agreement with the theoretical expectation.

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Complementarity is one of the essential paradigms of quantum mechanics [1]. Two quantities are mutually complementary in that complete (or partial) knowledge of one implies the complete (or partial) uncertainty about the other [2] and vice versa. The most generally known case is the complementarity between position and momentum, as expressed quantitatively in the Heisenberg uncertainty principle $\Delta x \Delta p \geqslant \hbar/2$. For neutrons the uncertainty relation has been demonstrated already back in 1966 by Shull [3]. Following the growing experimental efforts in atom optics during the last decade, the uncertainty principle has shown up implicitly in several experiments and has also been explicitly investigated in the time domain [4].

While being a physical phenomenon of interest in its own right, the complementarity between momentum and position is also an important factor for practical purposes: for example, it is applied for the preparation of transverse coherence in all experiments using collimated beams, a fact that can be mathematically phrased using the van Cittert-Zernike theorem [5–7].

There are good reasons to believe that complementarity and the uncertainty relation will hold, in principle, for all objects of the physical world and that these quantum properties are generally only hidden by technical noise for larger objects. It is therefore interesting to see how far this quantum-mechanical phenomenon can be experimentally extended to the macroscopic domain.

Here, we report on an experiment investigating in a quantitative way the uncertainty relation upon diffraction at a single slit for a molecule as complex, massive, and hot as the fullerene C_{70} ($m=840$ amu) emerging from the oven with an internal and translational temperature of 900 K.

It is well known that the limit $\hbar/2$ of the uncertainty relation $\Delta x \Delta p \geqslant \hbar/2$ is only reached for particular wave packets, for example, of the Gaussian-type. Evidently, the wave packet after passage through a rectangular slit is very different from this minimal uncertainty shape. This is also reflected in the far-field distribution that is described by the well-known sinc function rather than a Gaussian. It is therefore a matter of definition and convenience, which quantities to take as a measure of the position and momentum uncertainty in our case. Obviously, for a wave traversing a slit, one can take the slit width to be the measure of the spatial uncertainty Δx. The momentum uncertainty Δp can be related to the angular spread due to diffraction at the slit. Quantitatively, we define it as the momentum spread required to cover the full width at half maximum (FWHM) of the diffraction curve.

The setup of the experiment, shown in Fig. 1, is similar to that described in a previous publication [8]. An effusive thermal fullerene beam is produced at about 900 K. The velocity spread was as large as $\Delta v/v \sim 0.6$ and was taken into account in the numerical description of the experiment.

The molecular beam is collimated by two piezocontrolled slits. The width of the first slit S_1, is fixed at 10 μm, while the width Δx of the second slit S_2—which is located at the distance $L_1=113$ cm further downstream—can be varied to investigate the position-momentum uncertainty relation.

In order to also quantitatively describe the experiment the properties of the slits have to be known rather precisely. The slits (Piezosysteme Jena) are made of two silicon edges mounted on piezo controlled flexure stages. We obtain information about the slit opening in three different ways: from the applied piezovoltage, from the reading of a strain gauge mounted to the slits, and finally from the total number of molecules passing through the slit at a given opening. While the piezovoltage can be kept stable to better than $\Delta U/U < 10^{-4}$ it is well known that piezos show creep, hysteresis, and nonlinearities. However, it turned out in the experiments that the passive stability over a typical time of 1 h was of the order of 50 nm, as can be judged from the stability of the diffraction patterns. From a calibration of the hysteresis curve, we determine the change of the slit opening as a function of the piezovoltage. In order to know the absolute slit width we determined the zero position by measuring the number of molecules passing through the slit, when it was

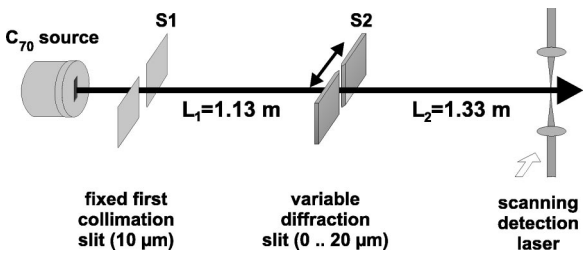

FIG. 1. Setup of the experiment. A thermal C_{70} beam is produced by sublimation of fullerene powder at 900 K. The beam is narrowed by S_1 and diffracted by S_2. S_1 is fixed at 10 μm. The width of slit S_2 is varied with ± 30 nm accuracy for $\Delta x < 1$ μm.

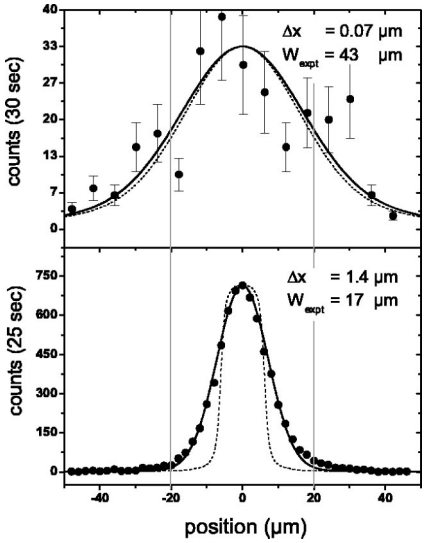

FIG. 2. Measured molecule distribution in the detection plane after passing through the piezocontrolled silicon slit S_2 having a width of $\Delta x = 1.4$ μm (bottom) and $\Delta x = 70$ nm (top). Both the quantum-mechanical calculation (continuous line) and the experiment (circle) show an increase of the beam width when going from medium (bottom) to narrow (top) slit widths. The dotted line indicates the wave calculation before the convolution with the known detector profile.

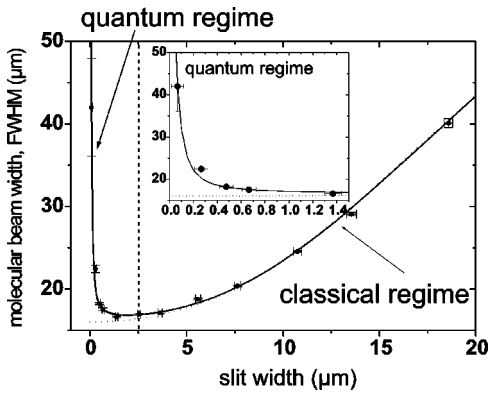

FIG. 3. Experimental molecular-beam width W_{expt} (full circle) is compared with the quantum prediction (continuous line) as a function of the slit opening Δx. The agreement is excellent across the whole range of slit openings (70 nm–20 μm). A purely classical shadow model predicts the dotted line and is in marked disagreement with the data for $\Delta x < 4$ μm. The latter is therefore designated as the quantum regime and magnified in the inset of Fig. 3.

being closed. We estimate this method to be accurate to within ± 30 nm.

We extract the momentum spread Δp after S_2 from the FWHM of the detected molecular beam W_{expt} in the detection plane, which is separated from S_2 by the length $L_2 = 133$ cm. The observed distribution function $f_{expt}(x) = D(x) \otimes M(x)$ is actually a convolution of the detector resolution function $D(x)$ and the real molecular-beam profile $M(x)$.

The scanning laser ionization detector has been characterized in depth in a previous publication [9]. For our present experiments with C_{70} the FWHM of the detector response was determined to be $D = 10 \pm 0.5$ μm at a laser power of $P = 10$ W. The effective FWHM detector height at this power was measured to be ~ 1 mm.

The second contribution, related to the measured molecular-beam profile $M(x)$, is composed of both the classical collimation and the momentum spread due to the quantum uncertainty. In order to compare the experiment with the uncertainty relation derived before, we concentrate in the following on the half-width values of these components only. Since the classical FWHM shadow width W_{cl} and the quantum contribution W_{qu} are completely independent their influence can be added quadratically to yield the FWHM value of $M(x)$, which we denominate as W_{expt}. The classical contribution W_{cl} can be derived from a simple geometrical shadow model. Taking the measured and the classically expected widths we can then deduce the contribution to the beam width due to the quantum uncertainty and we finally relate this spatial information to the corresponding momentum uncertainty, which then reads

$$\Delta p = \frac{p_z}{L_2}\{[(W_{expt})^2 - (W_{cl})^2]^{1/2} - \Delta x\}, \tag{1}$$

where p_z is the most probable longitudinal momentum of the molecules.

To trace out the uncertainty relation we varied the width of the second slit from about 20 μm down to roughly 50 nm and record the molecular-beam width in the detection plane. In Fig. 2, we show the measured molecular-beam profiles as full circles for two different widths of the second collimation slit. We see a relatively narrow beam of $W_{expt} = 17$ μm for the slit width $\Delta x = 1.4$ μm [Fig. 2(bottom)] and again a strong growth to $W_{expt} = 43$ μm for the slit width $\Delta x = (0.07 \pm 0.03)$ μm [Fig. 2(top)]. The error bars in Fig. 2 represent the statistical uncertainty due to the very low count rate, in particular at the smallest slit width.

The dashed line follows a full wave calculation as described below in order to show the molecular-beam profile as given by diffraction alone. The continuous curves represent the same model but convoluted with the detector profile.

Close inspection of the data shows a good agreement between the convoluted wave model and the experimental data. This good agreement is the first demonstration of single slit diffraction for a molecule as heavy, complex, and hot as C_{70}.

Figure 2(top) is actually an interesting complement to the high contrast interference fringes of fullerenes after diffraction at a nanofabricated grating with a grating constant of 100 nm, which we could demonstrate in a previous publication [10]. The single slit pattern shown here is the envelope of the far-field grating interference pattern. This provides a striking proof of the wave nature of the fullerene C_{70} because it demonstrates that the previous minima must have been due to destructive interference.

From the whole series of experiments with varying slit widths we have extracted the FWHM values from the experiment and we compare them with a quantum wave model in

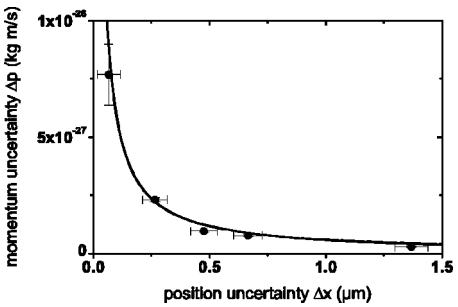

FIG. 4. Experimental verification of the Heisenberg uncertainty relation for C_{70}. The momentum uncertainty values Δp are derived from the far-field molecular-beam widths W_{expt} as described in the text. The position uncertainty Δx is given by the width of the second slit. The continuous line represents the expectation of a wave model for a monochromatic plane wave passing a slit.

Fig. 3. An excellent agreement between expectation and experiment is found throughout the whole range of values. We can distinguish essentially two different regimes corresponding to a pure quantum regime (left part of Fig. 3) and a range that can be very well described using a classical ball model (right part of Fig. 3). The continuous-wave calculation curve and the dotted classical line coincide almost completely down to a slit width of about $\Delta x = 4$ μm. Below this value the quantum-mechanical momentum spread Δp contributes significantly to the beam width in the detection plane. This quantum range is magnified in the inset of Fig. 3.

The horizontal error bars in this picture have two components, namely, the precision both of the absolute zero and of the scaling of the piezotranslation as a function of the applied voltage. Both are only important for a small slit width Δx. The absolute zero (closed) position of the piezoslits is known with an error of ± 30 nm, as mentioned above. The scaling with the applied piezo voltage is nonlinear and follows a hysteresis curve, which has been calibrated. We estimate an uncertainty of $\pm 3\%$ in the calibration of the hysteresis curve.

The vertical error bars estimate the uncertainty of the measured width of the beam in the detection plane. For small slit widths Δx these values are obtained from a least-squares Gaussian fit to the detected curve. For large Δx the marked trapezoidal shape as well as the high signal-to-noise ratio permit a direct reading of the experimental and theoretical FWHM values with very high accuracy.

The numerical simulations in Fig. 3 are based on the fact that the Schrödinger equation of our time-independent problem is formally equivalent to the Helmholtz equation and can, therefore, be treated using all the methods well known from optics. The solution is done in close analogy to the numerical approach as used in Ref. [11] for neutrons and similarly in Refs. [12,13] for atoms. There is no free parameter in the calculation except for a broadening of the detector resolution by 3.5 μm with respect to the best detector resolution curves recorded some time earlier [9]. This offset is most likely explained by a residual tilt of 2.7 mrad between laser and diffraction slit. This is in agreement with diffraction curves not shown here, which were recorded using the same setup but at half the width of the first collimation slit.

Since in previous papers it has been pointed out that the form factor of single slit diffraction may be influenced by the van der Waals interaction between the molecule and the slit walls [14,8], one may wonder whether this effect may become visible in the present experiment. However, the slit widths here, except for the smallest, are much larger than in the former grating diffraction experiments, where the effective slit width was reduced by about 15 nm. Since the van der Waals potential above a surface decreases with the third power of the object-wall distance the effect becomes small for the present study although the slit thickness is bigger than that of the previously used SiN_x gratings. For the smallest slit width $\Delta x \sim 70$ nm, a possible contribution is masked by the experimental error bar.

In the following, we compare our findings with the Heisenberg uncertainty relation between position and momentum. For this we use the method as indicated further above: From the measured beam width we separate the influence of the detector resolution in a deconvolution procedure. The remaining molecular-beam width is then decomposed into its classical and quantum part.

We can then plot Δp as derived from Eq. (1) as a function of Δx for slit openings lying well in the quantum regime and obtain Fig. 4. The full circles represent the values extracted from the experiment with error bars directly related to those of the inset of Fig. 3. The continuous line corresponds to the function $\Delta p = Ch/\Delta x$ with $C = 0.89$, which would be expected from diffraction theory for the passage of a monochromatic plane wave through a slit.

In conclusion, we regard the quantitative agreement between the experimental data and the predicted curve as a good support for the validity of the Heisenberg uncertainty principle for the fullerene C_{70}, i.e., for a system of interesting complexity. These experiments lend further support to our expectation that it will be possible to observe quantum interference phenomena for even larger molecules and clusters.

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