Comment on "Matter-Wave Interferometer for Large Molecules"

In a recent Letter [1] Brezgen et al. claim that a classical model of interactions is in principle unsuitable to reproduce interferometer experiments with C_{70} molecules, and that an optical model based on the de Broglie waves of the molecule explains the experimental data. In their Letter they clarify to a certain extent the objections raised by one of the authors [2] concerning their previous publication [3]. The clarification is much appreciated. However, it is not fully convincing.

To support their claim Brezgen et al. compute the visibility of the interference fringes based on two separate models: (i) A 'classical' model from geometrical optics, in which the movement of the molecules through a succession of optical gratings is described as rigid rays casting a shadow pattern at the detector plane; and (ii) a 'quantum' model, where the scalar potential of the grating leads to a phase shift of the de Broglie wave associated with the molecule. Additionally, the consequences of including a van der Waals term which gives a shift in scalar potentials is analyzed within both models.

These models are simplistic for various reasons. Most importantly, the main conclusion that the visibility of the interference fringes depends on the quantum character of the description is only based on the observation that the classical model does not have any term depending on the velocity of the molecules. But this is a deficiency of the model rather than something characteristic for the movement of molecules on classical trajectories. When the temperature of a molecular beam is increased, its changed velocity and translational energy will change its spatial distribution. This statistical effect has been analyzed in [5]. Fig. 1 (a) shows that it results in a velocity dependence of the visibility of the interference fringes of the earlier experiments in [3] within an entirely classical model as far as the movement of the atomic nuclei is concerned. This is no surprise for quantum chemists or theoretical solid state physicists, who are routinely using the Born-Oppenheimer approximation. In surface science or biochemistry such calculations have been successfully applied over the last decades to describe atomic movement with classical Lagrangians in the field of their electrons.

The description of van der Waals forces is problematic as well. In atom interferometry van der Waals forces are known to change the effective slit widths of diffraction gratings. But these descriptions can only be used as a good approximation for the long range forces, when there are no larger interaction terms. At high temperatures molecular vibrations lead to a fluctuating molecular dipole moment. We have computed the dipole moment of C_{60} molecules at the temperature of the experiments in [3] by first principles density functional theory (see Fig. 1 (b) [6]). As can be seen, the molecular dipole moment oscillates periodically along its path. These fluctuations are much larger in their amplitudes and time periods than the also time-dependent polarizations of the electronic shells of atoms responsible for van der Waals interactions. Whether the interactions of these fluctuating molecular dipoles with the diffraction gratings have a significant effect on the visibility of the interference fringes can only be answered, when a detailed model of the electronic structure of the surface of these gratings is provided.

While Brezgen et al. publish their visibility data, they do not publish another crucial information: the local distribution of maxima and minima with varying velocity. The omission does not help to make the paper convincing, because this is a decisive information for the discrimination between scattering and interference. If the velocity of the molecule is increased by 100%, then the number of visible interference peaks, given a constant grating, should be double the original number (see Fig. 2). It is difficult to obtain such a result, if the molecules are merely scattered.

![FIG. 1. (a) Simulation of the impacts of C_{60} molecules in a three slit system for narrow (solid line) and wide (dashed line) effective slit-widths. This figure exactly reproduces the intensity measurements in Ref. [3]. (b) Dipole moment of C_{60} molecules at 900 K calculated from first-principle density functional theory simulations.](image-a.png)

![FIG. 2. (a) Simulated interference pattern of a photon interferometer if the wavelength is equal to the distance between slits. (b) Simulated interference pattern if the wavelength is half the distance.](image-b.png)

We conclude that in order to really elucidate the physics behind these interference experiments with large...
molecules, an atomistic description of the molecular trajectories is needed, including all the interaction and excitation effects on their paths through the system, rather than just putting labels like 'classical' or 'quantum' on models based only on geometrical projections and wave propagation. We also conclude that crucial information is missing in the account of the experiments.

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