Pinning of quantized vortices in helium drops by dopant atoms and molecules

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Using a density functional method, we investigate the properties of liquid $^4$He droplets doped with atoms (Ne and Xe) and molecules (SF$_6$ and HCN). We consider the case of droplets having a quantized vortex pinned to the dopant. A liquid drop formula is proposed that accurately describes the total energy of the complex and allows one to extrapolate the density functional results to large $N$. For a given impurity, we find that the formation of a dopant+vortex+$^4$He$_N$ complex is energetically favored below a critical size $N_{cr}$. Our result support the possibility to observe quantized vortices in helium droplets by means of spectroscopic techniques.

Since the first observation of the $\nu_3$ vibrational band of SF$_6$ dissolved in $^4$He droplets [8], the infrared spectroscopy of molecules inside or attached to helium has attracted a wide interest (see, for instance, Refs. 8 and references therein). A major motivation for these efforts is that cold helium droplets offer the possibility of resolving rotational spectra of rather complex molecules and may constitute ‘the ultimate spectroscopic matrix’ 8 to create and study novel species. This unique feature of helium droplets originates from their quantum nature: not only they are fluid at zero temperature, due to the large zero point motion, but also exhibit a crucial superfluid behavior. The superfluid character of $^4$He droplets is interesting also from a fundamental viewpoint. In fact, the observation of superfluid effects in finite-sized quantum systems has to do with important concepts, like order parameter, Bose-Einstein condensation, and phase coherence, which were originally introduced for uniform systems and which are now widely used in different contexts.

In the case of liquid helium, Grebenev et al. 9 recently showed that only a rather small amount of $^4$He atoms is needed to develop a superfluid droplet, also confirming theoretical predictions 8. In that experiment, the evidence for superfluidity is the appearance of a sharp rotational spectrum of an OCS molecule in $^3$He-$^4$He mixed drops, when the number of $^4$He atoms surrounding the dopant 8 is larger than about 60. In the same spirit, experiments have been made to observe critical velocities 8 (i.e., the occurrence of a Landau criterion for superfluidity), and a reduction of the moment of inertia (see 11 and references therein). In contradistinction, detecting quantized vortices in droplets still remains an open question. It is worth stressing that all these investigations have many analogies with the current activity on Bose-Einstein condensation in trapped gases, where new results are now available about critical velocities 11, moments of inertia 12,13 and vortices 14.

In this work we address the problem of quantized vortices. One first observes that a vortex line in a pure droplet is expected to be difficult to produce and stabilize, since it implies a significant increase of energy compared to a vortex-free droplet. In order to circumvent this limitation, we explore the possibility of pinning the vortex line to a dopant atom or molecule. If the dopant is deeply bound inside the droplet, it might stabilize the vortex for a time long enough to permit its observation. A second advantage is that the dopant could make the detection feasible via spectroscopic techniques.

Our purpose is to determine the energy and density profile of a impurity+vortex+$^4$He$_N$ complex, for droplets up to $N = 1000$, using a finite-range density functional. We then subtract to its energy that of the same droplet without vortex and/or impurity and show that the difference fits very well to a liquid drop formula, which allows one to safely extrapolate to larger droplets. The density functional method consists in minimizing the total energy of the system at zero temperature written as a functional of the He density. We use the Orsay-Paris functional 15, which is based on an effective non-local interaction with a few parameters fixed to reproduce known properties of bulk liquid He. This functional has been shown to accurately reproduce the static properties of pure and doped He clusters 16 and has also been used to describe a quantized vortex line in bulk liquid helium 17. In the latter case, the vortex is included with the Feynman-Onsager (OF) ansatz for the velocity field. This implies a singular vorticity and hence the vanishing of the density on the vortex axis. At $T = 0$ this approximation is a reasonable starting point, since it enormously reduces the computational cost. Recent calculations 18 have shown that the density profile and energy of the vortex line given by the OF approximation are reasonably close to the ones.
obtained by assuming non-singular vorticity. Finally, the actual temperature of the droplets, below 0.4 K \([2]\), is low enough for neglecting thermal contributions to the fluid motion. Thus, corrections beyond OF and at \(T \neq 0\) are expected not to change the main results of the present work.

The minimization of the energy is performed in axial symmetry by mapping the density on a grid of points, putting the vortex line along the \(z\)-axis and the dopant in the center, at \(r = 0\). The numerical code used to calculate the density profile and energy is the same used in \([14]\). The potentials for rare gas impurities have been taken from \([12]\), that of the spherically averaged SF\(_6\) from \([20]\), and that of HCN from \([23]\).

We first consider pure droplets with and without vortex. In Fig. \(2\) we show density profiles, at \(z = 0\), obtained for different \(N\). For large droplets the shape approaches that of a rectilinear vortex in the uniform liquid \([17]\); the core radius is of the order of \(1 \div 2\) Å and the density oscillates as a consequence of the He-He interaction. In Fig. \(2\) we plot the energy associated to the vortex flow, defined as

\[
\Delta E_V(N) = E_V(N) - E(N)
\]

where \(E_V\) and \(E\) are the energies of droplets with and without vortex, respectively. The solid line represents the results obtained with a liquid drop formula of the kind:

\[
\Delta E_V(N) = \alpha N^{1/3} + \beta N^{1/3} \log N + \gamma N^{-1/3},
\]

with the parameters \(\alpha = 2.868 \text{ K}, \beta = 1.445 \text{ K}, \text{ and } \gamma = 0.313 \text{ K}\) extracted from a fit to the density functional calculations. This formula works well. The reason can be easily understood by means of a hollow-core model for the vortex, having core radius \(a\), in a droplet of radius \(R\) and constant density \(\rho_0\). By integrating the kinetic energy of the vortex flow in the limit \(R \gg a\), one gets

\[
E_{\text{kin}} = \frac{2\pi \hbar^2 \rho_0}{m_4} \left[R \log \left(\frac{2R}{a}\right) - R + \frac{a^2}{4R}\right].
\]

Writing \(R = r_0 N^{1/3}\) one recovers the \(N\)-dependence as in \([16]\).

The next step is the inclusion of a dopant atom or molecule. As an example, in Fig. \(3\) we show the He density distribution for a drop of \(N = 500\) with HCN hosted in the vortex core. Both the axis of the linear molecule and that of the vortex are taken along \(z\). The density is very inhomogeneous near the dopant, due to the complexity of the HCN-He interaction. The energetics of the system can be conveniently analysed by introducing the following energies:

\[
\Delta E^X_V(N) \equiv E_{X+V}(N) - E_X(N),
\]

\[
S_X(N) \equiv E_X(N) - E(N),
\]

\[
S_{X+V}(N) \equiv E_{X+V}(N) - E(N),
\]

where the subscripts \(X\) and \(V\) refer to drops doped with impurity \(X\) and/or vortex line.

The energy \(\Delta E^X_V\) is the one associated with the vortex flow in the doped cluster. In Fig. \(2\) it is compared with the vortex energy in pure droplets, \(\Delta E_V\). The difference

\[
\delta_X(N) = \Delta E^X_V(N) - \Delta E_V < 0
\]

is almost independent of \(N\), apart from the smallest droplets. The reason is that this difference has to do with the ‘geometrical extension’ of the dopant, i.e., the ‘hole’ made by the dopant in the vortex flow, as well as with the distortion of the density near the dopant caused by the pinning of the vortex core. Both effects are localized near the dopant and, thus, they are expected to give a shift in energy which becomes \(N\)-independent for large droplets.

The quantity \(S_X(N)\) in Eq. \(2\) is the solvation energy of the dopant in a vortex-free droplet. The results obtained for Ne, Xe, HCN and SF\(_6\) are shown in Fig. \(1\). As already discussed in \([16,22]\), the solvation energy becomes almost \(N\)-independent for \(N\) larger than a few hundreds. The value at \(N = 1000\) can be safely taken to represent the solvation energy in the bulk, \(S_X(\infty) \approx S_X(1000)\). For our analysis, we have chosen impurities having binding energies on a wide range.

The key quantity in the present study is the solvation energy of the dopant+vortex complex given by \(S_{X+V}(N)\) in Eq. \(1\). The results are shown in Fig. \(2\). From the definitions \(1\) to \(6\) one can also write

\[
S_{X+V}(N) = E_X(N) + \Delta E^X_V(N) - E(N) = S_X(N) + \Delta E_V(N) + \delta_X(N).
\]

In Fig. \(2\) we compare \(S_{X+V}\) with the sum \(S_X + \Delta E_V\); the difference is \(\delta_X\). The simple picture which emerges from this analysis is that the solvation energy of the dopant+vortex complex is just the sum of the solvation energy of the dopant with no vortex and the extra energy of a vortex in a pure droplet, apart from a small shift which depends on the dopant. Deviations from this rule are significant only for small droplets, having radius of the order of the size of the dopant. Our numerical results provide a quantitative basis for this picture and yield typical estimates of \(\delta_X\). It is worth noticing that, by rearranging the terms in \(2\), this quantity can be written as the difference between the solvation energies of the dopant in a droplet with an without vortex,

\[
\delta_X = [(E_{X+V} - E_V) - (E_X - E)],
\]

and can hence be interpreted as the binding energy of the dopant to the vortex \([23]\).

Since the solvation energy \(S_X\) is negative and almost constant for \(N > 300\) while the vortex energy \(\Delta E_V\) always increases, the dopant+vortex complex has a solvation energy which changes sign at some \(N_{\alpha}\). This means that for \(N < N_{\alpha}\), the dopant+vortex complex is energetically favored. In the case of Ne, as one can see in
Fig. 1. $N_\text{cr} \sim 380$. This number is rather small as compared to the typical droplet size in current experiments, and is a consequence of the weak binding of Ne. Dopants with stronger binding have larger $N_\text{cr}$. An estimate for HCN, Xe, and SF$_6$ can be easily obtained by means of the liquid drop formula. One has to insert expression (3) in (8) and use the large-$N$ values of $S_X$ and $\delta_X$. The $S_X$ values turn out to be $-310$ K, $-320$ K and $-622$ K, and the $\delta_X$ values are 5.0 K, 4.4 K, and 7.7 K, for Xe, HCN, and SF$_6$, respectively. These numbers yield $N_\text{cr} \approx 7600$ for Xe, $\approx 8100$ for HCN, and $\approx 40000$ for SF$_6$.

In conclusion, the analysis of the energetics of doped helium droplets has allowed us to disclose a possible mechanism to create and stabilize vortex lines. A dopant has a large solvation energy. A metastable state appears attached to the dopant, since the binding energy $\delta_X$ is negative. The formation of the complex is energetically favored below a critical $N$ which is well within the range of droplet sizes met in current experiments if the dopant has a large solvation energy. A metastable state could also exist for $N > N_\text{cr}$, but estimating its lifetime is a more demanding calculation. One should also explore the energy barrier associated with other possible decay processes. Further work is planned in this direction.

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