Coexistence of vortex arrays and surface capillary waves in spinning prolate superfluid $^4$He nanodroplets

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Abstract

Within Density Functional Theory, we have studied the interplay between vortex arrays and capillary waves in spinning prolate $^4$He droplets made of several thousands of helium atoms. Surface capillary waves are ubiquitous in prolate superfluid $^4$He droplets and, depending on the size and angular momentum of the droplet, they may coexist with vortex arrays. We have found that the equilibrium configuration of small prolate droplets is vortex-free, evolving towards vortex-hosting as the droplet size increases. This result is in agreement with a recent experiment [S. M. O. O’Connell et al., Phys. Rev. Lett. 124, 215301 (2020)], where it has been disclosed that vortex arrays and capillary waves coexist in the equilibrium configuration of very large drops. Contrarily to viscous droplets executing rigid body rotation, the stability phase diagram of spinning $^4$He droplets cannot be universally described in terms of dimensionless angular momentum and angular velocity variables: instead, the rotational properties of superfluid helium droplets display a clear dependence on the droplet size and the number of vortices they host.

I. INTRODUCTION

Superfluid $^4$He droplets produced in the expansion of a cold helium gas or by hydrodynamic instability of a liquid helium jet passing through the nozzle of a molecular beam apparatus have been considered the ultimate inert matrix for molecular spectroscopy, constituting also an ideal testground to study superfluidity at the nanoscale. A series of experiments aiming at determining the appearance of large spinning $^4$He droplets made of $10^8 - 10^{11}$ atoms has motivated a renewed interest in aspects such as how vortices are distributed inside helium droplets, how impurities are captured by the vortex lines they host, and how impurities arrange inside vortex-hosting droplets.

Two experimental techniques have been used to address large spinning $^4$He droplets. The pioneering work of Ref. [1] and other works carried out by the same group [2,3] used coherent diffractive imaging of x-rays from a free electron laser (FEL) and gave access to a model-independent determination of the two-dimensional (2D) projection of the droplet density on a plane perpendicular to the x-ray incident direction via iterative phase retrieval algorithms; by doping He droplets with Xe atoms they were also able to detect the presence of vortex arrays. In Refs. [4,5] He droplets were irradiated with intense extreme ultraviolet pulses (XUV), and the measurements of wide-angle diffraction patterns provided access to full three-dimensional information. However, it was not possible to directly determine the shapes of the droplets, and so the analysis was carried out parameterizing the droplet density with a combination of two ellipsoidal caps connected by a hyperboloidal center-piece, and all their defining parameters were determined by matching the experimental diffraction patterns with those obtained from simulations.

These remarkable experiments have shown that most helium droplets are spherical and only a few percent are deformed. In particular, the analysis carried out by Langbehn et al. on a sample consisting of a large number of droplets (38,150) showed that 92.9% of them were spherical, 5.6% were spheroidal (oblate), and 1.5% were prolate. The latter population was made of ellipsoidal (0.8%), pill-shaped (0.6%), and dumbbell-like (0.1%) droplets, where the appearance was quantitatively determined by evaluating the distance of the center of mass of the droplet to its surface along the direction of the principal axes of inertia. The results obtained with both experimental techniques were compared to calculations made for incompressible viscous droplets subject to surface tension and centrifugal forces, it was concluded that they were in agreement.

The precise meaning of this agreement needs some clarification, as otherwise one might be left with the impression that the rotational properties of superfluid $^4$He and of classical rotating viscous fluids are very similar, and this is not the case. The experiments have indeed shown that the shapes of spinning superfluid $^4$He drops are the same as those found for viscous drops subject to centrifugal and surface tension forces, and that some relationships between ratios of the mentioned distances are in agreement with the classical results, see, e.g., Fig. 3 of Ref. [6] and Fig. 1 below. However, to ascertain whether the sequence of shapes is the same, which is what determines the equilibrium phase diagram, it is necessary to experimentally determine the angular mo-
momentum for a large enough sample of droplets; this has not been possible yet. Only very recently, the angular momentum and shape of a few He drops have been simultaneously determined.\textsuperscript{15}

At first glance, the mere fact that oblate \(^{4}\)He spinning drops have been identified\textsuperscript{12,16,18} is surprising and unexpected. Large helium drops are produced in the normal phase and may acquire angular momentum during the passage of the fluid through the nozzle of the molecular beam apparatus. The drops cool down to a temperature of 0.37 K\textsuperscript{19} and are thus superfluid when they are probed. The natural question is how a spinning superfluid drop may acquire an oblate (axisymmetric) shape as this is quantum mechanically forbidden. In fact the equations characterizing the macroscopic behaviour of a superfluid at zero temperature have the form of irrotational hydrodynamics, from which the moment of inertia can be calculated as \(\Theta_{ir} = \varepsilon^2 \Theta_{rig}\), where

\[
\varepsilon = \frac{\langle y^2 - x^2 \rangle}{\langle y^2 + x^2 \rangle}
\]

and \(\Theta_{rig} = Nm(x^2 + y^2)\) is the rigid moment of inertia, \(N\) being the number of atoms in the droplet and \(m\) the atomic mass, showing that in a superfluid the value of the moment of inertia is smaller than the rigid value. In particular, for axisymmetric systems (i.e. \(\langle x^2 \rangle = \langle y^2 \rangle\)) the above relation gives for the angular momentum of the superfluid \(L_z = \Theta_{ir} \omega = 0\). Notice that these results are not restricted to the perturbative regime of small \(\omega\).\textsuperscript{28} These expressions show that vortex-free oblate axisymmetric configurations cannot spin, whereas prolate (non axisymmetric) configurations can, and the resulting angular momentum \(L_{cap} = \Theta_{ir} \omega\) is associated with the so-called capillary waves (see the discussion in the following).

Thus, the only likely explanation for the experimental observation of spinning oblate \(^{4}\)He droplets is that these drops contain a vortex array which can store most of the angular momentum acquired during the passage through the experimental apparatus—some is taken away by the atoms emitted during the cooling process. It has indeed been shown\textsuperscript{12,29} that the presence of a large enough vortex array in the \(^{4}\)He droplets confers to them a globally oblate appearance\textsuperscript{29} similarly to the case of rotating viscous droplets. Vortex arrays have been experimentally detected only in a few cases\textsuperscript{12,29,30} as they escape detection unless drops are doped and the presence of vortices is established by the appearance of Bragg patterns from the impurities that fill the vortex cores.

The existence of helium drops that follow the sequence of prolate shapes characteristic of viscous drops under the effect of rotation is also worth discussing. In this case, axial symmetry is spontaneously broken \((\langle x^2 \rangle \neq \langle y^2 \rangle)\), and the droplet can store a finite amount of angular momentum in the form of capillary waves, even in the absence of vortices. In general, however, capillary waves and vortices may coexist in spinning prolate droplets, producing two quite different irrotational velocity flows compatible with the superfluidity requirement: one associated to quantized vortices, and the other to surface capillary waves.\textsuperscript{35,36} Remarkably, in the stability phase diagram, spinning prolate superfluid drops whose angular momentum is exclusively stored in capillary waves lie on a line\textsuperscript{20} quite distinct from the classical line\textsuperscript{29} as discussed in the following.

There is another unexpected difference between a rotational viscous fluid and an irrotational superfluid with finite angular momentum. When a spherical viscous droplet is set into rotation, as a consequence of centrifugal forces it flattens out in the direction of the rotational axis: the shortest half-axis of the droplet \((c_z,\text{ as defined in the following})\) is thus aligned with the rotational axis. However, this is not always the case for irrotational flows: spinning vortex-free droplets with prolate shapes have instead the intermediate half-axis \((b_y,\text{ as defined in the following})\) aligned with the rotational axis.\textsuperscript{33} When vortices are present, in most cases the shortest half-axis \(c_z\) is again aligned with the rotational axis, in agreement with the classical behavior. Consequently, if only the shape of the droplet is experimentally identified, the rotational axis cannot be determined unambiguously if no further information about the angular momentum vector is available.

From the above discussion it appears that capillary waves and vortex arrays can coexist in prolate superfluid drops. This has been demonstrated in a recent combined experimental and Density Functional Theory (DFT) work\textsuperscript{18} The DFT analysis was restricted to deformable self-sustained superfluid \(^{4}\)He cylinders which could host a fairly large vortex array due to the lower dimensionality (2D) of the computational real space grid. This model was a generalization of the rotating, elliptic cylindric vessel filled with \(^{4}\)He studied in Ref.\textsuperscript{49}

In this work we complement the discussion made in Ref.\textsuperscript{18} by addressing several prolate configurations corresponding to large (for DFT standards) superfluid \(^{4}\)He droplets: we put the emphasis on the interplay between vortices and capillary waves, we determine the equilibrium configuration for two representative values of the angular momentum, and we discuss the similarities and differences between superfluid (irrotational) and viscous (rotational) behavior. Oblate droplets hosting a different number of vortices have been thoroughly studied in Ref.\textsuperscript{19}

This paper is organized as follows: the DFT approach to liquid helium is recalled in Sec.\textsuperscript{1} Sec.\textsuperscript{11} presents the results and their discussion; and a summary is given in Sec.\textsuperscript{14} Multimedia information is presented in the Supplemental Material\textsuperscript{18}

## II. THEORETICAL APPROACH

Density Functional Theory in its static and time-dependent (TDDFT) versions has proved to be a pow-
The above equation has been solved using the 4He-DFT-BCN-TLS computing package\textsuperscript{23} see Refs.\textsuperscript{12} and\textsuperscript{15} and references therein for additional details. Briefly, we work in cartesian coordinates, with the effective wave function $\Psi(r)$ defined at the nodes of a 3D grid inside a calculation box large enough to accommodate the droplet in such a way that the He density is sensibly zero on the box surface. Periodic boundary conditions are imposed which allow to use the Fast Fourier Transform to carry out the convolution needed to obtain the DFT mean field $\mathcal{H}[\rho]$. The differential operators in $\mathcal{H}[\rho]$ and $\hat{L}_z$ are approximated by 13-point formulas and the typical space step is 0.8 Å. Working at fixed angular momentum requires to iterate on the value of $\omega$; there are efficient ways of adjusting iteratively $\omega$, e.g., the Augmented Lagrangian method\textsuperscript{26}.

Angular momentum can be stored in a superfluid $^4$He droplet in the form of surface capillary waves and/or quantized vortices\textsuperscript{18,20,23} In order to deposit angular momentum of both types in the droplet, we have used the so-called “imprinting” procedure by starting the imaginary time minimization from a very flexible guess for the effective wave function $\Psi(r)$, namely a superposition of a quadrupolar capillary wave and $n_v$ vortex lines parallel to the $z$ axis,

$$
\Psi_{0}(r) = \rho_0^{1/2}(r) e^{i\alpha xy} \prod_{j=1}^{n_v} \frac{(x-x_j) + i(y-y_j)}{\sqrt{(x-x_j)^2 + (y-y_j)^2}},
$$

which is then optimized by iteratively solving Eq. (5). Here, $\rho_0(r)$ is an arbitrary, vortex-free droplet density, the complex phase $e^{i\alpha xy}$ imprints a surface capillary wave with quadrupolar symmetry around the $z$ axis, and the product term imprints a vortex array made of $n_v$ linear vortices\textsuperscript{22} where $(x_j, y_j)$ is the initial position of the $j$-vortex core. The initial value of $\alpha$ and the vortex core positions are guessed, and during the iterative solution of Eq. (5) both the vortex core structure and positions, and the droplet shape change to provide at convergence the lowest energy configuration.

Writing $\Psi(r) \equiv \phi(r) \exp[i\mathcal{S}(r)]$, the velocity field of the superfluid is

$$
v(r) = \frac{\hbar}{m} \text{Im} \left\{ \frac{\nabla \Psi(r)}{\Psi(r)} \right\} = \frac{\hbar}{m} \nabla \mathcal{S}(r)
$$

The velocity potential $\mathcal{S}(r)$ gets contributions from both capillary waves and vortices in a complex way, and their contributions cannot be rigorously disentangled —the analysis needs to be model-dependent.\textsuperscript{18}

The existence of different prolate configurations for a $^4$He$_N$ droplet spinning at a given $L$, each of them characterized by a number of vortices $n_v$, raises the question of which is the globally stable configuration and how does it depend on the number of atoms in the droplet. To determine which is the globally stable configuration among different ones, one has to compare their total energy $E[\rho]$.
 TABLE IV: Energetics and morphology of prolate \(^4\)He droplets with \(N = 35,000\) at \(\Lambda = 0.80\) for \(n_v = 0 - 6\). \(L\) indicates a linear, \(C\) a cross-shaped, and \(P\) a pentagonal vortex array. The 6\((Ct)\) configuration has a center vortex, \(i.e.,\) along the rotational axis. The entry in boldface corresponds to the globally stable configuration, and the asterisk identifies the highest energy configuration among those with the same \(n_v\) value; in particular, 6\(^*\) corresponds to a 6-vortex array configuration in which the six vortices are at the vertices of a hexagon.

| \(n_v\) | \(E\) (\(\text{K}\)) | \(E_{\text{kin}}\) (\(\text{K}\)) | \(\omega \) (\(\text{K}/\text{b}\)) | \(a_x\) (\(\text{Å}\)) | \(b_y\) (\(\text{Å}\)) | \(c_z\) (\(\text{Å}\)) |
|-------|----------------|-----------------|-----------------|----------------|----------------|----------------|
| 0     | -227.393.50   | 3360.11         | 0.02402         | 120.49         | 45.42          | 54.21          |
| 1     | -227.522.50   | 3581.02         | 0.02580         | 116.29         | 49.68          | 54.09          |
| 2     | -227.629.67   | 3767.38         | 0.02697         | 111.55         | 52.80          | 55.29          |
| 3     | -227.698.13   | 3986.17         | 0.02789         | 106.56         | 56.28          | 53.89          |
| 4\((C)\)|-227.721.40   | 4233.29         | 0.02927         | 101.38         | 61.12          | 56.03          |
| 4\((L)\)|-227.700.04   | 4231.98         | 0.02829         | 101.76         | 60.15          | 55.82          |
| 5\((P)\)|-227.723.95   | 4476.43         | 0.03009         | 95.16          | 66.16          | 56.86          |
| 6\((Ct)\)|-227.699.01  | 4732.32         | 0.03130         | 85.12          | 75.48          | 53.36          |
| 6\(^*\)|-227.697.29   | 4710.33         | 0.03012         | 87.06          | 73.60          | 57.59          |

TABLE III: Energetics and morphology of prolate \(^4\)He droplets with \(N = 35,000\) at \(\Lambda = 1.25\) for \(n_v = 0 - 5\). \(L\) indicates a linear, \(C\) a cross-shaped vortex array. The entry in boldface corresponds to the globally stable configuration, and the asterisk identifies the highest energy \(n_v\) \(= 4\) configuration.

| \(n_v\) | \(E\) (\(\text{K}\)) | \(E_{\text{kin}}\) (\(\text{K}\)) | \(\omega \) (\(\text{K}/\text{b}\)) | \(a_x\) (\(\text{Å}\)) | \(b_y\) (\(\text{Å}\)) | \(c_z\) (\(\text{Å}\)) |
|-------|----------------|-----------------|-----------------|----------------|----------------|----------------|
| 0     | -225315.35    | 4174.71         | 0.02102         | 137.65         | 38.98          | 43.97          |
| 1     | -225378.53    | 4450.00         | 0.02225         | 134.88         | 42.80          | 43.50          |
| 2     | -225323.77    | 4695.91         | 0.02318         | 131.14         | 44.40          | 46.30          |
| 3     | -225312.04    | 4950.83         | 0.02402         | 127.81         | 47.07          | 46.14          |
| 4\((C)\)|-225239.14   | 5198.51         | 0.02477         | 125.33         | 50.06          | 46.70          |
| 4\((L)\)|-225263.35   | 5217.47         | 0.02464         | 124.69         | 49.46          | 47.12          |
| 5\((P)\)|-225185.14   | 5466.69         | 0.02529         | 122.15         | 52.15          | 47.86          |
| 5\(^*\)|-225173.77   | 5489.12         | 0.02505         | 122.16         | 51.69          | 45.90          |

TABLE V: Energetics and morphology of prolate \(^4\)He\(_{1500}\) droplets at \(\Lambda = 1.25\) for \(n_v = 0 - 4\). \(L\) indicates a linear vortex array. The entry in boldface corresponds to the globally stable configuration.

| \(n_v\) | \(E\) (\(\text{K}\)) | \(E_{\text{kin}}\) (\(\text{K}\)) | \(\omega \) (\(\text{K}/\text{b}\)) | \(a_x\) (\(\text{Å}\)) | \(b_y\) (\(\text{Å}\)) | \(c_z\) (\(\text{Å}\)) |
|-------|----------------|-----------------|-----------------|----------------|----------------|----------------|
| 0     | -225693,40    | 4692.54         | 0.02600         | 118.84         | 54.80          | 48.43          |

TABLE VI: Energetics and morphology of prolate \(^4\)He droplets with \(N = 35,000\) at \(\Lambda = 1.25\) for \(n_v = 0 - 5\). \(L\) indicates a linear, \(C\) a cross-shaped, and \(P\) a pentagonal vortex array. The 6-vortex array configuration is a stretched hexagon. The entry in boldface corresponds to the globally stable configuration, and the asterisk identifies the highest energy configuration among those with the same \(n_v\) value.

Including the rotational energy (Routhian) \(^{[29,30]}\) at variance with the classical case, where a rotational energy term in the rigid body approximation has to be added to the energy of the droplet \(^{[29,31]}\) in the DFT approach to superfluid \(^4\)He in the corotating frame this is naturally accounted for through the velocity field embodied in the phase of the effective wave function \(\Psi(\mathbf{r})\), see Eq. \(^7\), so one does not need to add any extra term to the total energy expression, Eq. \(^2\) nor to Eq. \(^5\).

A difficulty inherent to the study of helium droplets is that configurations with rather distinct morphologies may have similar energies; therefore, a careful analysis is required in order to distinguish the global minimum from metastable configurations. An added challenge is that most vortex array configurations are very robust and, in spite of not being a physically conserved quantity, the starting \(n_v\) value is often conserved instead of being relaxed to the optimal value corresponding to the global equilibrium configuration. This forces to explore, for a given \(N\)-atoms droplet, all possible \(n_v\) values compatible with the chosen \(L\) —a task which becomes increasingly cumbersome as \(L\) increases. This robustness has however the benefit that one has access to a series of excited configurations, which might be experimentally accessible, characterized not only by the strictly conserved \(N\) and \(L\) values but also by the number of vortices \(n_v\). We have always checked the stability of the vortex array against shape distortions.

For every stationary configuration obtained by solving Eq. \(^3\), a “sharp density surface” is determined by calculating the locus at which the helium density equals \(\rho_0/2\), where \(\rho_0\) is the atom density of the liquid; for a spherical distribution this corresponds to the sphere of radius \(R\) defined below. In the case of deformed droplets, three lengths (half-axes) \(a_x\), \(b_y\) and \(c_z\) are introduced representing the distances from the center of mass of the droplet to the sharp surface along the principal axes of inertia. These lengths are represented in the inset in Fig. \(^[1]\). For an axisymmetric droplet, \(a_x = b_y \neq c_z\). These lengths have been used to characterize the droplet shape by defining two dimensionless aspect ratios, \(a_x/c_z\) and \(b_y/V^{1/3}\), where \(V\) is the volume of the non-rotating spherical droplet.
The shapes of classical drops exclusively subject to surface tension and centrifugal forces follow a universal stability diagram in terms of the dimensionless angular momentum $\Lambda$ and angular velocity $\Omega$ defined by

\begin{align}
\Omega &= \sqrt{\frac{m \rho_0 R^3}{8 \gamma}} \omega \\
\Lambda &= \frac{h}{\sqrt{8 \gamma R^3 m \rho_0}} L
\end{align}

where $m$ is the atom mass and $\gamma$ is the surface tension of the liquid. For liquid $^4$He at zero temperature and pressure, $\gamma = 0.274$ K $\text{Å}^{-2}$, $\rho_0 = 0.0218$ $\text{Å}^{-3}$, the sharp radius of the spherical droplet is given by $R = r_0 N^{1/3}$ with $r_0 = 2.22$ $\text{Å}$ and $\hbar^2/m = 12.12$ K $\text{Å}^2$. We recall that $\hbar = 7.6382$ K ps and that $\hbar c = 2.2899 \times 10^7$ K $\text{Å}$.

We want to stress that this universality is lost if one goes beyond that simple model, e.g., by using a DFT description of the liquid, since this incorporates effects such as surface diffuseness and liquid incompressibility although such effects are expected to be less important as the droplet size increases, they can be tangible for the droplet sizes investigated here. Yet, Eqs. (8) are very useful as they allow for a comparison between experimental and calculated droplets, even if the latter are much smaller (for obvious computational reasons) than those probed in the experiments. However, one should have in
FIG. 3: Densities on the symmetry plane perpendicular to the rotational axis for the $N = 35,000$ He droplet hosting zero, one, and six vortices at $\Lambda = 0.80$. The streamlines of the superfluid flow are superimposed and their direction is counterclockwise. The positions of the vortex cores are marked by red dots for visualization. The color bar shows the atom density in units of $\text{Å}^{-3}$, and the black bar represents a distance of 100 Å.

mind that some differences may appear in the comparison due to these unavoidable finite-size effects. Besides, the presence of vortices in the case of superfluid droplets definitely breaks the universality of the stability diagram, as shown below.

### III. RESULTS AND DISCUSSION

Systematic DFT calculations are very cumbersome to carry out, especially when several configurations hosting different numbers of vortices have to be analyzed and their geometries have to be tested against distortions of the vortex array. This hinders a systematic exploration of the phase diagram. For this reason, we have limited our study to some relevant cases which embody the physics we aim to discuss. We have focused in particular on two $\Lambda$ values that embrace a fairly large range of prolate configurations, namely $\Lambda = 0.80$ and 1.25, for droplets with $N = 1500$ atoms (a value for which a rather detailed series of DFT calculations is available) and with $N = 20,000$ and 35,000; these large prolate droplets (for DFT standards) are well suited to disclose the interplay between capillary waves and vortex arrays object of this study, and how the number of vortices in the globally stable configuration evolves as a function of $N$.

Tables II–VI collect the results obtained for the mentioned $(\Lambda, N)$ values (one pair of values per table) and give details on their energetics and morphology. In each table we have written in boldface the entry corresponding to the globally stable configuration, i.e., that with the lowest energy for the given values of $N$ and $\Lambda$. When
more than one configuration has been found for a given number of vortices \( n_v \), the more energetic one is identified with an asterisk (*). Triangular, cross-shaped, pentagonal and linear vortex arrays are denoted by \( T, C, P, \) and \( L \), respectively. Notice that the configurations in a given table can be directly compared, as they correspond to the same values of two strictly conserved quantities in isolated droplets.

As already discussed in the Introduction Section, it appears from the Tables that for vortex-free superfluid droplets the intermediate half-axis \( b_y \) is aligned with the rotational axis, at variance with the classical rotating droplets where the shortest half-axis \( c_z \) is aligned instead with the rotational axis. We are not aware of a general demonstration of this counter-intuitive result: its proof is however known for ellipsoidal drops made of irrotational (potential) fluids.\(^{33}\) This peculiarity of spinning irrotational drops has to be taken into account for a proper analysis of the experiments: identifying the shortest axis of a spinning superfluid \(^4\)He droplet with the rotational axis might be incorrect. For vortex-hosting superfluid droplets, though, the rotational axis coincides most of the times with the shortest \( (c_z) \) one, as for viscous drops. But there are exceptions, \( e.g. \), for \( N = 20,000 \) and 35,000 when \( n_v = 2 \).

In most cases, the presence of a vortex array confers to the droplet the appearance of a rotating viscous droplet. A notorious example of this apparently classical behavior is the meniscus that develops, at the liquid-vapor interface, in a rotating bucket filled with superfluid helium above the critical angular velocity required for vortex nucleation.\(^{37,48}\) Hence, determining the angular momentum of the droplet (magnitude and direction) and whether it hosts a vortex array or not seems unavoidable before drawing a definite conclusion about how superfluid helium droplets rotate.

We show in Fig. 1 the dimensionless ratios \( b_y/V \) versus \( a_x/c_z \) for the configurations collected in the tables, irrespective of whether they correspond to globally stable or metastable configurations. The \( N = 1500 \) results have been complemented with those shown in Fig. 1 of Ref. 21 which presents data for a large sample of \( \Lambda \) values. Also shown are the experimental results,\(^{13} \) which have an average atom number \( \langle N \rangle = 6 \times 10^9 \), and the result for classical rotating drops.\(^{32} \) While the results for vortex-hosting configurations are similarly distributed and roughly follow the classical drops trend, the figure shows that vortex-free prolate configurations form a separate branch (capillary branch) displaying a completely different behavior. Both branches meet at the \((a_x/c_z, b_y/V) = (1,3/4\pi)\) point corresponding to spherical droplets.

It is worth noticing that configurations with the same \( n_v \) but rather different \( N \) values yield similar points on the \((a_x/c_z, b_y/V)\) plane, especially when the vortex array has the same morphology (linear, cross, ...) and in particular for vortex-free configurations. The universality of the classical results, which allows to scale them with \( N \), is not completely lost. Unfortunately, even \( N = 35,000 \) is too small a value compared to the experimental ones to allow us to draw any sensible conclusion about this issue. The figure also shows that, for a fixed \( \Lambda \) value, the larger the \( n_v \), the more similar viscous and vortex-hosting droplets are.

We thus see that knowledge of the shape of a large number of deformed configurations is not enough to unambiguously characterize their rotational behavior as classical or superfluid, and that one has to combine this information with the simultaneous determination of the angular momentum of the droplet by, \( e.g. \), obtaining the stability diagram in the \((\Lambda,\Omega)\) plane. This diagram is shown in Fig. 2 for some of the configurations in Fig. 1. While the capillary branch may arrive up to nearly \( \Lambda = 0 \), as only a small deformation is needed for the droplet to sustain capillary waves, the vortex-hosting branch must abruptly interrupt at some \((\Lambda_c,\Omega_c)\) point, as a critical angular velocity \( \omega_c \) is needed for nucleating a vortex line in a superfluid droplet.\(^{30}\) The red symbols in Fig. 2 represent the critical vortex nucleation point for the three \( N \) values used in this study. For the sake of completeness, we also display the DFT result for \( N = 5000 \)\(^{30}\) and that of the hollow core mode\(^{33}\) for a \( N = 10^8 \) drop taking for the vortex core radius a value of 1A.

The existence of this critical point dramatically distinguishes superfluid from viscous droplet rotational behavior, as no axisymmetric superfluid He droplet can be set into rotation for \( \Lambda < \Lambda_c \). Another conspicuous difference between viscous and superfluid vortex-hosting droplet results can be observed in Fig. 2, namely the location of the oblate-to-prolate bifurcation point. For viscous droplets, it is at \( \Lambda = 1.2 \)\(^{30,31}\) whereas for droplets of the size studied here, DFT yields the bifurcation slightly below \( \Lambda = 0.8 \). Indeed, the three-vortex \(^4\)He\(_{1500}\) configurations connected by a red horizontal line correspond to oblate (left) and a prolate (right) configurations. We attribute this large difference to a finite size effect, although we cannot ascertain this. In Fig. 2 finite size effects also clearly affect the position of the critical vortex nucleation point.

Comparing the results for the same \( N \) value, one can see from the tables that the ratio \( b_y/a_x \) decreases as \( \Lambda \) increases. This causes, \( e.g. \), that for the \(^4\)He\(_{1500}\) droplet, the most stable \( n_v = 4 \) vortex array, that was cross-shaped at \( \Lambda = 0.80 \), is instead linear at \( \Lambda = 1.25 \); the same happens for \( N = 20,000 \). Large droplets can host both kinds of vortex arrangements, \( e.g. \), Table V1. For a given \( \Lambda \), a large droplet may accommodate a non-linear vortex array more easily. Notice for instance that, at \( \Lambda = 1.25 \), the cross-shaped \( n_v = 4 \) configuration for \( N = 35,000 \) is still more stable than the linear one. The existence of multiple configurations for the same \( n_v \) value is also ubiquitous in axisymmetric configurations,\(^{19,20,31}\) making the search for the lowest energy configuration very time-consuming.

Further inspection of the tables shows that, for given \( N \) and \( \Lambda \) values, the globally stable spinning prolate configu-
FIG. 4: Densities on the symmetry plane perpendicular to the rotational axis for the $N = 35,000$ He droplet hosting zero, one, and $n_v = 3 - 6$ vortices at $\Lambda = 1.25$. The streamlines of the superfluid flow are superimposed and their direction is counterclockwise. The positions of the vortex cores are marked by red dots for visualization. The color bar shows the atom density in units of $\text{Å}^{-3}$, and the black bar represents a distance of 100 Å.

Droplets are not necessarily vortex-free, and that, contrary to a naive expectation, for large, fixed $N$ and $\Lambda$ values, the energy is not monotonously varying with $n_v$, see for instance Tables I, II and VI. We have found that for the largest $\Lambda$ value considered in this study ($\Lambda = 1.25$), the vortex-hosting configuration becomes the globally stable configuration only above a “critical” $N$ value. We have checked that for $N = 30,000$ the vortex-free and the $n_v = 2$ configurations are nearly degenerate with energies of $-191,994.86$ K and $-191,994.49$ K, respectively. As Table VI shows, the globally stable configuration of the $N = 35,000$ droplet is the $n_v = 2$ one. It is thus very likely that the prolate large $N$ drops studied in the FEL and XUV experiments contain both vortex arrays and capillary waves. Notice also that the energy differences between the first excited configurations and the globally stable ones are small, and it cannot be completely discarded that some of such excited configurations are also detected in the experiments. These considerations indicate that vortex-free prolate configurations might be extremely difficult to detect either because they correspond to globally stable configurations only for small-$N$ droplets that escape detection in current diffractive imaging experiments, or because they are metastable configurations in the case of large-$N$ drops and likely decay to vortex-hosting configurations before they are probed.

Figures 3 and 4 show some 2D densities corresponding to the $N = 35,000$ droplet. One may see that, in order to store a large angular momentum, a vortex-free configuration has to be very stretched –see the discussion on capillary waves below. This is a general trend that implies an increase of the droplet surface energy; as shown in the tables, the kinetic energy also increases with the number of vortices. The globally stable configuration is determined by a delicate balance between these two competing effects. Figure 4 shows that the $N = 35,000$ droplet is large enough to accommodate up to 6 vortices, and that the shape of the droplet evolves from two-lobed to a more compact, “pill-shaped” appearance as found in the experiments. Despite most of these configurations are metastable, these shape transitions exemplify the competition between compact and more linear vortex arrays arrangements to determine the appearance of the droplet. Notice that Fig. 4 also dis-
plays the same effect: as $n_v$ increases for a given $\Lambda$, the point representing the vortex-hosting configuration dramatically displaces to smaller $a_z/c_z$ values, yielding at the same time a larger $b_y^3/V$, hence more compact droplet shapes. It is also worth seeing from the 2D figures that two-lobed configurations as those reported in Refs. 14 and 16 appear when $n_v$ is small; increasing $n_v$ reduces the wrist between the lobes, as the vortex array becomes less linear and more similar to a patch of a triangular arrangement (as expected when $n_v$ becomes very large). Since the number of pill-shaped droplets found in the experiment\[16\] is six times larger than that of dumbbell-like droplets, this would again indicate that large, very prolate droplets host fairly large vortex arrays.

Figure 5 displays a side view of the $N = 35,000$ droplet at $\Lambda = 1.25$ for the $n_v = 0-3$ configurations. One can see how vortices locally distort the droplet surface as they have to hit it perpendicularly. A similar figure can be found in Ref. 19 for oblate configurations. Thus, droplets hosting large vortex arrays are far from being ellipsoidal. In the diffractive imaging experiments, the images used to identify the presence of vortices with a well oriented direction of their cores\[18\] would correspond to the flat surfaces limiting from above and below the droplet side view shown in this figure.

Vortex arrays in prolate configurations are distorted, as found in the experiments\[15\] and displayed in the 2D figures. To characterize these distortions, we have calculated an oblate configuration “twin” of the prolate $N = 1500$, $\Lambda = 0.80$, $n_v = 3$ configuration for the same value of $\Omega$, namely 0.5247, which implies a lower $\Lambda$ value, see Fig. 2. For this $n_v = 3$ oblate configuration we have found $\Lambda = 0.714$, $a_z/c_z = 1.38$, and $b_y^3/V = 0.318$, the latter two values to be compared to those of its twin prolate configuration, $a_z/c_z = 1.68$ and $b_y^3/V = 0.207$. In the oblate $n_v = 3$ configuration the vortex array forms an equilateral triangle with an inter-vortex distance of 16.6 Å. In the $n_v = 3$ prolate configuration the triangle is isosceles and stretched in the direction of the largest principal axis of inertia ($x$ axis). The stretched intervortex distance is 22.1 Å, and the other two sides of the triangle correspond to an inter-vortex distance of 15.4 Å.

All configurations displayed in the 2D figures are stationary in the corotating frame; as a consequence, they would be seen from the laboratory frame as if they were rotating, apparently like a rigid-body, with the angular frequency $\omega$ imposed to obtain them. Let us recall and stress that this is a misleading appearance. The motion of a fluid is a combination of translation, rotation and deformation of the fluid elements, and only when the vorticity (defined as he curl of the velocity field of the fluid) is non-zero, one may speak of a true rotation, being important to distinguish between rotation and motion of the fluid elements along a curved path, e.g., around a vortex core. Since the velocity field of the superfluid is potential, vorticity is zero except on the vortex lines; in their absence, vorticity is zero everywhere.

To illustrate this situation, we have simulated within the TDDFT approach\[22\] the time evolution of the prolate $^4\text{He}_{1500}$ droplet for the vortex-free and the $n_v = 3$ configurations at $\Lambda = 0.8$. Fig. 6 shows, for the $n_v = 3$ case, the trajectory $(x_c, y_c)$ of the vortex cores on the $z = 0$ plane. It can be observed that the inter-vortex distances do not change during the evolution, indicating that the three-vortex array rotates with the angular velocity imposed to the corotating frame, $\omega = 1.871 \times 10^{-2} \text{ ps}^{-1}$. We see this dynamics as the rigid rotation of the vortex cores (which is not in contradiction with the previous discussion, as they are empty), around which the irrotational superflow accommodates in its displacement. One may also notice the accuracy of the stationary so-

FIG. 5: Densities on the symmetry plane containing the vortex lines for the $N = 35,000$ droplet hosting $n_v = 0-3$ vortices at $\Lambda = 1.25$. The color bar shows the atom density in units of Å$^{-3}$, and the black bar represents a distance of 100 Å.

FIG. 6: Trajectories of the vortex cores $(x_c, y_c)$ on the $z = 0$ plane corresponding to the prolate $N = 1500$, $\Lambda = 0.80$, $n_v = 3$ configuration. One vortex is turning around the center of mass of the droplet (shown by a cross) at a distance of 7.7 Å, while the other two vortices rotate at a distance of 11.0 Å. The inset represents the droplet density on the symmetry plane perpendicular to the rotational axis.
lution we have obtained; otherwise, the vortex core trajectories, plotted here point by point, would not be the perfect circumferences displayed in Fig. 3. The figure also exemplifies the robustness of the fixed-Ne configurations mentioned in Sec. II. These configurations—which are stationary in the corotating frame—likely correspond to local energy minima and are separated by an energy barrier from other configurations having a different Ne but the same N and Λ values.

The interested reader may find two videos in the Supplemental Material showing the time evolution of these two configurations in the corotating frame for about 450 ps. It may be illuminating to look at the time evolution of the streamlines pattern: it shows how different the apparent rotation of the superfluid droplet is, compared to the true rotation of a viscous drop. We want to stress that the energy, particle number and angular momentum of the isolated droplet are strictly conserved during the evolution.

It is worth recalling that vorticity in quantum fluids may often lead to a turbulent regime, like, e.g., that resulting from the decay of vortex tangles in bulk liquid 4He, or the turbulence in Bose Einstein condensates (BEC) generated by fast stirring the condensate using laser beams. Our vortex structures, however, are stable against decay, reconnections or other mechanisms which are known to lead to turbulent behavior in liquid 4He or BECs. This is what we indeed observe in our simulations: in spite of the possibility of bending and displacing of the vortex cores, which is allowed by the three-dimensional nature of our calculations, the vortex structures we find are either stationary states in the co-rotating system or, when let to evolve under real-time dynamics, they rotate around the central axis but remain otherwise stable, without showing any tendency to be expelled or reconnect with nearby vortices. This is in agreement with the experimental measurements on nanoscopic 4He droplets hosting vortices where no sign of decay of the vortex structures is observed. Turbulent behavior may arise though in nanoscopic 4He droplets under special conditions, as illustrated in Ref. 55, where a TD-DFT study of the merging of two vortex-free superfluid 4He droplets has shown that vortex rings are dynamically nucleated during the merging process and that their annihilation produces a massive emission of rotons and subsequent turbulent behavior, whose scaling follows the very general power laws underlying turbulence in quantum fluid systems.

Streamlines of the superfluid flow are shown in Figs. 4 and 5. The superflow follows the direction of the angular frequency imposed to the corotating frame (counterclockwise in this case). Streamlines allow to infer by visual inspection the coexistence of vortices and surface capillary waves, as their velocity fields are very different. The streamlines associated to vortices wrap around their cores, whereas those associated to capillary waves end abruptly at the droplet surface. As already mentioned, separating the contribution of capillary waves and vortices in the velocity fields is generally not possible, the superfluid velocity field being proportional to the gradient of the phase $\mathbf{S}(\mathbf{r})$ of the effective wave function $\Psi(\mathbf{r})$, where the contributions of both vortices and capillary waves are entangled.

The coexistence of one single vortex and a quadrupolar irrotational flow in an elliptic cylinder filled with helium rotating at constant $\omega$ was studied by Fetter, who anticipated the possible appearance of a row of vortices along the longer axis of the elliptic vessel cross section. At variance with isolated droplets, the cylinder has a rigid surface of known geometry. Writing the phase of $\Psi(\mathbf{r})$ as the sum of a term arising from the vortex line and another from the quadrupolar flow (the same guess that we made in Eq. 6, but in our case only for the starting configuration), he could determine how the angular momentum is shared between the vortex and the quadrupolar irrotational flow. It does not look feasible to extend the cylinder approach to the $n_v > 1$ case. Even for one single vortex, the study of isolated droplets is much more complex, as their shapes are not ellipsoidal in general and they are only known after the stationary configuration has been determined by solving Eq. 6.

Yet, to analyze the experimental results it has been useful to know, at least approximately, how angular momentum is shared between vortices and capillary waves. In order to do that, we computed the deformation parameter $\varepsilon$ defined in Eq. 1 using the $a_x$ and $b_y$ parameters given in the tables for the $n_v = 0$ configurations, and the relation giving the angular momentum associated to capillary waves, $L_{\text{cap}} = \varepsilon^2 \Theta_{\text{rig}} \omega$ (see the discussion following Eq. 1).

We have found that it works surprisingly well, as it coincides with the exact $L$ calculated within the DFT approach to within a few percent. The $L_{\text{cap}}$ value obtained for each studied $n_v = 0$ configuration is given in the tables. For vortex-hosting configurations, the angular momentum stored in the vortex arrays may then be estimated subtracting $L_{\text{cap}}$ from the total $L$. The validity of this procedure cannot be assessed though. So far, the only possibility to experimentally determine the angular momentum of the drop has been to use Feynman’s formula to obtain the angular velocity $\omega$ from the vortex density, which could be determined from the 2D droplet images, and the above expression for $L_{\text{cap}}$, see Ref. 18 for details.

IV. SUMMARY

We have shown that the presence of vortex arrays in spinning superfluid 4He droplets and their interplay with capillary waves has a profound influence in the determination of their globally stable configuration. The presence of vortex arrays, irrespective of whether they are detected or not, is the only plausible explanation for the existence of oblate configurations, which otherwise will be in conflict with quantum mechanics or would im-
ply that droplets are not superfluid, which is extremely unlikely due to the working temperatures in $^4$He droplets experiments.$^3$

Surface capillary waves are ubiquitous in prolate superfluid $^4$He droplets. Depending on their size and angular momentum, these waves may coexist with vortex arrays. We have found that the global equilibrium configuration of small prolate droplets is vortex-free, but the situation changes as the droplet size increases. This result is in agreement with a recent experiment$^4$ where it has been disclosed that vortex arrays and capillary waves coexist in very large drops.

While vortex arrays may be detected by doping the droplets, capillary waves are very elusive and escape direct detection. This poses a serious difficulty to determine the angular momentum of the superfluid droplet. From the theoretical side, microscopic approaches such as DFT treat vortex arrays and capillary waves on the same footing, and one has to resort to approximate methods to disentangle their contribution to the total angular momentum of the droplet. Determining –even in a model dependent way– the angular momentum of the droplets is essential if one wants to characterize spinning superfluid droplets, as only simultaneous knowledge of the droplet morphology and angular momentum allows for a sensible comparison with classical or quantum models.

Contrarily to viscous droplets executing rigid body rotation (modeled by viscous droplets subject to surface tension and centrifugal forces alone), the stability phase diagram of superfluid droplets is not universal and cannot be characterized by dimensionless angular momentum $\Lambda$ and dimensionless angular velocity $\Omega$ as its classical counterpart. Their knowledge does not determine univocally the rotational properties of superfluid helium droplets, which display a clear dependence on the droplet size and/or the number of vortices they host. This is not in contradiction with the recent finding$^5$ that big superfluid $^4$He droplets hosting a large number of vortices seem to rotate like rigid-bodies, thus following the classical stability phase diagram. Rather, it is a manifestation of finite size effects which still have to be studied in detail and, together with recent works on rotating $^3$He droplets$^6$ and mixed $^3$He-$^4$He droplets$^7$ call for further experimental research.

Acknowledgments

We are indebted to Andrey Vilesov for useful discussions. We thank Thomas Möller and Bruno Langbehn for providing us with the results of Ref. 16 shown in Fig. 1 and Sam Butler for providing us with the results of the classical model calculations used in this work. This work has been performed under Grant No FIS2017-87801-P (AEI/FEDER, UE). J.M.E. acknowledges support from the Spanish Research Agency (AEI) through the Severo Ochoa Centres of Excellence programme (grant SEV-2017-0706) and from the European Union MaX Center of Excellence (EU-H2020 Grant No. 824143).

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