

# Nanoscopic jets and filaments of Helium at zero temperature:

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# Nanoscopic jets and filaments of superfluid $^4\text{He}$ at zero temperature: A DFT study

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## ABSTRACT

The instability of a cryogenic  $^4\text{He}$  jet exiting through a small nozzle into vacuum leads to the formation of  $^4\text{He}$  drops, which are considered ideal matrices for spectroscopic studies of embedded atoms and molecules. Here, we present a He-density functional theory (DFT) description of droplet formation resulting from jet breaking and contraction of superfluid  $^4\text{He}$  filaments. Whereas the fragmentation of long jets closely follows the predictions of linear theory for inviscid fluids, leading to droplet trains interspersed with smaller satellite droplets, the contraction of filaments with an aspect ratio larger than a threshold value leads to the nucleation of vortex rings, which hinder their breakup into droplets.

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## I. INTRODUCTION

Liquid  $^4\text{He}$  droplets at low temperature offer a unique environment for molecular spectroscopy<sup>1–3</sup> and the study of superfluidity on the atomic scale,<sup>4–6</sup> including the study of quantum vortices.<sup>7–10</sup> Usually,  $^4\text{He}$  droplets are produced by the expansion of cooled  $^4\text{He}$  gas or by the instability of a cryogenic  $^4\text{He}$  jet exiting a source chamber into a vacuum throughout a nozzle, whose temperature and pressure determine the appearance of the liquid jet and the droplet size and velocity distributions.<sup>11,12</sup>  $^4\text{He}$  drops undergo evaporative cooling and become superfluid, eventually reaching a temperature of 0.4 K<sup>11</sup> on a  $\mu\text{s}$  time scale.<sup>13</sup>

Understanding the dynamical properties of liquid  $^4\text{He}$  jets and the instabilities leading to their fragmentation is a relevant issue in the production and characterization of droplets made of  $^4\text{He}$ . This unique fluid allows for a large variation of non-dimensional parameters related to the fluid viscosity and the velocity at which it exits the nozzle, which characterize its dynamical properties.<sup>14</sup> This understanding also has a primary application, namely to make available  $^4\text{He}$  drops with the size and velocity required by the experiments, together with size and velocity distributions as narrow as possible. This has led to recent experimental studies on the disintegration of liquid  $^4\text{He}$  jets.<sup>15,16</sup> Besides, a liquid thread with a

finite length (“filament” in the following) and no external constraint is expected to contract, trying to minimize its surface energy, and eventually reach a spherical liquid drop. However, the outcome of the process is not always that simple, as an ample body of experiments and theoretical work on classical fluids has shown over the years. We notice that liquid  $^4\text{He}$  filaments are regularly found in the experiments.<sup>14–16</sup>

Liquid jets and filaments and their dynamical instabilities are well established subjects of study in classical fluid dynamics because of the practical questions and applications on the one hand and because jet dynamics probes many physical properties and theoretical approaches on the other hand (see, e.g., Refs. 17–19 and references therein). Most studies concentrate on viscous fluids because of their practical implications. The underlying theoretical and numerical challenge is to solve the Navier–Stokes (NS) equation subject to appropriate boundary conditions.

The effect of viscosity and surface tension is embodied in the Ohnesorge number  $Oh$ , defined as  $Oh = \mu / \sqrt{m\rho_0\gamma R_0}$ , where  $m$  is the atom mass,  $\rho_0$  is the atom density of the fluid,  $\gamma$  is the surface tension,  $\mu$  is the viscosity coefficient, and  $R_0$  is the radius of the jet or filament. Inviscid filaments have been addressed in passing by Schulkes,<sup>20</sup> but owing to computational challenges, he could not simulate the extreme interfacial deformations arising in crucial

moments of the dynamics, as during filament breaking and end-pinching, i.e., the formation of two isolated drops from the opposite tips of the filaments. While it is naturally assumed that when solving the NS equation for small enough viscosities the results should be nearly indistinguishable from the inviscid limit, see, e.g., Refs. 19 and 21, a description of superfluid (i.e., inviscid and irrotational) jets and filaments is not available in the literature. Such a study may be of relevance in view of the aforementioned studies on superfluid  $^4\text{He}$  droplets, which is the motivation of the present work. Our goal is to describe, at the microscopic level, the dynamics of contraction and breaking of zero temperature superfluid  $^4\text{He}$  nanojets and nanofilaments in vacuum using the well-established  $^4\text{He}$  density functional theory (He-DFT) approach.<sup>22–25</sup> The He-DFT approach is similar, in the superfluid  $^4\text{He}$  phase, to the Gross–Pitaevskii approach, which has successfully been applied to the description of cold gases in the superfluid Bose–Einstein condensate phase, in particular in the study of quantized vortices.<sup>26–28</sup>

Helium density functional and time-dependent density functional (He-TDDFT) methods have proven to be very powerful tools to address superfluid  $^4\text{He}$  samples. Within the He-DFT approach, the finite range of the helium–helium van der Waals (vdW) interaction is explicitly incorporated in the simulations. As a consequence, the liquid–vacuum interface has a non-zero surface width, which is important in the description of nanoscopic  $^4\text{He}$  systems like the jets and filaments studied in the present work. It also takes into account the finite compressibility of the fluid, so the possibility of having density excitations (ripples, phonons, and rotons) is naturally incorporated into the simulations. It also considers the possibility of atom evaporation from the  $^4\text{He}$  sample during the real-time dynamics,<sup>29</sup> which, however, has been found to be a negligible effect in the present study.

The He-DFT approach adds to the classical viscous fluids or molecular dynamics descriptions the possibility of disclosing purely superfluid effects in the dynamics, in particular quantized vortex nucleation. It has been recognized that a retracting viscous liquid filament may escape from end-pinching through the creation of vortex rings for Ohnesorge numbers in the  $0.002 < Oh < 0.1$  range.<sup>21</sup> Here, we show that the same happens in the zero viscosity, irrotational superfluid case.

Due to the computational burden associated with fully three-dimensional He-DFT simulations such as the ones discussed here, we address jets and filaments of nanoscopic size. Studies on the breakup of liquid nanojets are available in the literature, and atomistic molecular dynamics simulations on the formation, stability, and breakup of viscous fluids have been carried out.<sup>30</sup> To our knowledge, no simulations of the breakup of superfluid nanojets and filaments have been published so far.

This work is organized as follows. In Sec. II, we briefly present the He-DFT approach used in this work. In Sec. III A, we discuss the results for the dynamics of  $^4\text{He}$  jets, focusing on the conditions leading to fragmentation, and in Sec. III B, we study the contraction and possible break-up of  $^4\text{He}$  filaments with finite length. A summary with some concluding remarks is presented in Sec. IV. In addition to the main text, we provide in the [supplementary material](#) the real-time dynamics of the  $^4\text{He}$  jets and filaments addressed in this paper. This multimedia material constitutes an important part of this work since it helps capture physical details that would otherwise escape the written account.

## II. THEORETICAL APPROACH

Density functional theory for liquid helium is a phenomenological approach that constitutes a good compromise between accuracy and feasibility. The parameters of the functional have been adjusted to reproduce various properties of the bulk superfluid, such as equilibrium density, energy per atom, and compressibility, as well as the main features of the dispersion relation of the elementary excitations of superfluid  $^4\text{He}$ .<sup>22</sup> A detailed description of the method can be found in Refs. 23–25.

Within He-DFT, the energy of a  $N$ -atom sample is written as a functional of the  $^4\text{He}$  atom density  $\rho(\mathbf{r})$  as

$$E[\rho] = T[\rho] + E_c[\rho] = \frac{\hbar^2}{2m} \int d\mathbf{r} |\nabla\Psi(\mathbf{r})|^2 + \int d\mathbf{r} \mathcal{E}_c[\rho], \quad (1)$$

where the first term is the kinetic energy,  $m$  is the mass of the  $^4\text{He}$  atom, and  $\Psi(\mathbf{r})$  is the effective wave function (or order parameter) of the superfluid such that  $\rho(\mathbf{r}) = |\Psi(\mathbf{r})|^2$  with  $\int d\mathbf{r} |\Psi(\mathbf{r})|^2 = N$ . The functional  $\mathcal{E}_c(\rho)$  we have used contains the He–He interaction term within the Hartree approximation and additional terms describing non-local correlation effects.<sup>31</sup>

The equilibrium configuration of the system is obtained by solving, using an imaginary-time relaxation method,<sup>25</sup> the Euler–Lagrange equation

$$\left\{ -\frac{\hbar^2}{2m} \nabla^2 + \frac{\delta\mathcal{E}_c}{\delta\rho} \right\} \Psi \equiv \mathcal{H}[\rho] \Psi = \zeta \Psi, \quad (2)$$

where  $\zeta$  is the  $^4\text{He}$  chemical potential corresponding to the number of He atoms in the sample.

Minimizing the action associated with Eq. (1) leads to the He-TDDFT equation

$$i\hbar \frac{\partial\Psi}{\partial t} = \left\{ -\frac{\hbar^2}{2m} \nabla^2 + \frac{\delta\mathcal{E}_c}{\delta\rho} \right\} \Psi \equiv \mathcal{H}[\rho] \Psi, \quad (3)$$

from which one can simulate the real-time evolution of the system.

The above equations have been solved using the  $^4\text{He}$ -DFT-BCN-TLS computing package<sup>32</sup> (see Refs. 24 and 25 and references therein for additional details). Briefly, we work in cartesian coordinates, with the effective wave function  $\Psi(\mathbf{r}, t)$  defined at the nodes of a 3D grid inside a calculation box. Periodic boundary conditions (PBC) are imposed, which allow to use the fast Fourier transform<sup>33</sup> to efficiently compute the convolutions needed to obtain the DFT mean field  $\mathcal{H}[\rho]$ . The differential operators in  $\mathcal{H}[\rho]$  are approximated by 13-point formulas. Equations (2) and (3) have been solved using a space-step of 1.2 Å, and the time-dependent Eq. (3) has been numerically integrated using a Hamming predictor-modifier-corrector initiated by a fourth-order Runge-Kutta-Gill algorithm<sup>34</sup> with a time-step of 2 fs. This time-step has been found to keep the energy of the jet and filaments properly conserved during the dynamics, as it corresponds to non-dissipative processes. We have also checked that the jet configurations obtained in the course of the dynamics are robust against reasonable changes in the chosen space-step.

### III. RESULTS

We have considered jets and filaments of sharp radius  $R_0 = 21.5$  Å, defined as the radius at which the density equals  $\rho_0/2$ , with  $\rho_0$  being the liquid  $^4\text{He}$  atom density at zero temperature and pressure.

#### A. $^4\text{He}$ nanoscopic jet

The physics of classical liquid jets has been reviewed by Eggers and Villermaux.<sup>17</sup> The thinning and breakup of a liquid jet is mainly determined by surface tension effects. The stability of an infinite fluid cylinder of radius  $R_0$  was studied by Plateau,<sup>35</sup> who showed that it exists in an unstable equilibrium and that any perturbation with wavelength  $\lambda$  greater than  $2\pi R_0$  is unstable and allows the surface tension to break up the cylinder into droplets, thus decreasing the surface energy of the system. Rayleigh later showed<sup>36</sup> that for an inviscid liquid, the fastest growing mode occurs when the wavelength of the axial undulation that ultimately leads to the fragmentation of the jet into droplets is equal to  $\lambda_c = 9.01R_0$  (Rayleigh–Plateau instability). When the jet breaks up, one or more small satellite drops—resulting from the necks breaking—may form between the larger droplets.

The characteristic times for jet instability and breakup are set by the capillary time  $\tau_c$  defined as

$$\tau_c = \sqrt{\frac{m\rho_0 R_0^3}{\gamma}}, \quad (4)$$

with  $\gamma$  being the surface tension of the liquid. In the case of  $^4\text{He}$ , we have  $m = 4.325 \times 10^{13}$  K,  $\rho_0 = 0.021836$  Å<sup>-3</sup>, and  $\gamma = 0.274$  K Å<sup>-2</sup>. Hence,  $\tau_c(R_0 = 21.5$  Å) = 61.7 ps.

It is customary to define the aspect ratio as  $\Gamma = \tilde{L}/R_0$ , where  $\tilde{L} = L/2$  is the half-length of the jet. Here,  $L$  coincides with the length of the simulation cell in the jet direction. From the linearized fluid dynamics equations for an inviscid and incompressible fluid, a critical value  $\Gamma_c$  is predicted to trigger jet fragmentation.<sup>36</sup> It corresponds to the mode with wavelength  $\lambda_c = 2\pi/k$ , where  $k$  is such that  $\omega(k)$  is the maximum. Here<sup>17</sup>

$$\omega^2(k) = \left[ \xi \frac{I_1(\xi)}{I_0(\xi)} (1 - \xi^2) \right] \frac{1}{\tau_c^2}, \quad (5)$$

where  $I_0(x)$  and  $I_1(x) = dI_0(x)/dx$  are modified Bessel functions of the first kind and  $\xi \equiv kR_0$ . From the maximum of  $\omega$ , one finds  $kR_0 = 0.697$  and thus

$$\Gamma_c = \pi/0.697 = 4.505. \quad (6)$$

In correspondence with it one has

$$\omega_{\max} = 0.343 \sqrt{\gamma/(m\rho_0 R_0^3)} = \frac{0.343}{\tau_c}. \quad (7)$$

Similarly to what occurs in classical liquid jets, we have shown that the He-TDDFT approach yields the Rayleigh–Plateau instability for the superfluid nanojet when it is subject to a perturbation with the right wavelength. Since the evolution takes place in vacuum, i.e., in the absence of ambient gas embedding the jet, the velocity of the jet itself is not playing any role and, therefore, we perform our simulations in a reference frame where the jet is at rest (comoving frame).

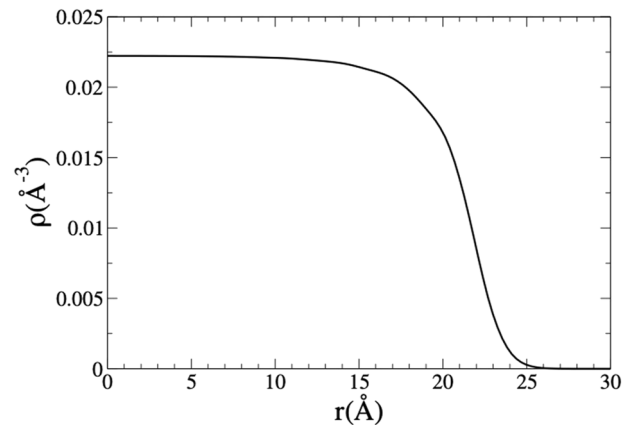


FIG. 1. Density profile in the radial direction of a cylinder of radius  $R_0 = 21.5$  Å representing a  $^4\text{He}$  nanojet.

To this end, we simulate the jet with a cylindrical filament in a simulation box subject to PBC along the cylinder axis (the  $x$ -axis in the following). Its equilibrium structure has been obtained by solving Eq. (2); a plot of the jet density profile in the transverse direction is shown in Fig. 1. The jet displays a bulk region of fairly constant density (slightly higher than the bulk  $^4\text{He}$  density  $\rho_0$  due to the compressive effect exerted by the surface tension on the lateral surface of the cylinder), delimited by a surface with a finite width. As mentioned earlier, the radius of the cylinder  $R_0$  is defined as the distance from the symmetry axis of the point where  $\rho(r) = \rho_0/2$ .

We have verified by imaginary-time dynamics that the cylinder is indeed unstable against a small initial axial perturbation with the proper wavelength. To do so, we consider a (periodically repeated) cylinder made of  $N = 12076$   $^4\text{He}$  atoms and length  $L = 387.2$  Å. We have first found the equilibrium geometry with a resulting radius  $R_0 = 21.5$  Å. Therefore, the aspect ratio of the cylinder is  $\Gamma = \tilde{L}/R_0 = 9.01$ , i.e., twice the critical aspect ratio Eq. (6); in this way, the axial undulation caused by the mode with the Rayleigh–Plateau wavelength  $\lambda_c$  will produce two necks along the jet, inducing fragmentation into two droplets, as shown in the following.

Next, we performed an imaginary-time dynamics starting from a configuration corresponding to a slightly perturbed axially symmetric cylinder of radius  $R_0$ , where the initial density profile is given by

$$\rho(\mathbf{r}) = \frac{\rho_0}{\exp\left\{\left[\sqrt{y^2 + z^2} - R(x)\right]/0.5\right\} + 1}, \quad (8)$$

with

$$R(x) = R_0[1 - \varepsilon \cos(4\pi x/L)], \quad (9)$$

and  $\varepsilon \ll 1$ . With our choice of the length  $L$  and radius  $R_0$ , the wavelength of the resulting density modulation is precisely equal to  $\lambda_c = 9.01R_0$ . The form in Eq. (9) ensures that the perturbed density is normalized to have the same number of atoms as the unperturbed cylinder. If  $\delta_0 = \varepsilon R_0$ , the maximum excursion of the radius along the cylinder axis is thus  $R = R_0 \pm \delta_0$ .

The total energy of the axially perturbed cylinder turns out to be lower than that of the unperturbed cylinder, i.e., the system is energetically unstable toward a deformation leading to fragmentation. Starting from this configuration, we have performed an imaginary-time relaxation during which two necks develop, eventually leading, as they shrink to zero, to two identical spherical droplets as the lowest energy state.

Next, we have studied, by solving the He-TDDFT Eq. (3), the actual real-time dynamics of the fragmentation process, starting from the axially perturbed cylindrical jet. Following Ref. 37, the perturbation is applied both to the density, as described earlier, and to the axial velocity of the jet as well, using the linearized solution of the Rayleigh–Plateau instability,

$$v = v_0 \sin(4\pi x/L), \quad (10)$$

where  $v_0 = 2\delta_0 v_{\max}/R_0$ . Notice that the radial perturbation is symmetric at the origin, whereas the velocity fluctuation is antisymmetric. Here,  $v_{\max} = \omega_{\max} R_0/\xi_{\max}$  is calculated from Eq. (5) using  $\xi = \xi_{\max} = 0.697$ , giving  $v_{\max} \sim 17$  m/s. Our starting value for the perturbation amplitude is  $\delta_0 = 0.452$  Å, corresponding to the choice  $\varepsilon = 0.021$  in Eq. (9).

In order to apply this velocity field to the superfluid jet, we multiply the initial axially perturbed cylinder wave function  $\Psi(\mathbf{r}) = \rho^{1/2}(\mathbf{r})$  by the phase  $e^{i\phi}$  with

$$\phi = -2 \frac{\delta_0}{R_0} v_{\max} \left( \frac{L/2}{2\pi} \right) \cos(4\pi x/L), \quad (11)$$

and proceed with the real-time evolution.

Figure 2 shows snapshots of the jet density on a symmetry plane containing the cylinder axis during the real-time dynamics. It can be seen that, starting from the perturbed cylinder, undulations whose amplitude increases with time appear along the jet. The

instability is caused by the fact that Laplace pressure increases in constricted regions, driving out the fluid and thereby reducing the neck radius further. The jet evolves into density bulges connected by thin threads. Threads eventually break up, and isolated drops appear instead. Figure 2 also shows that the threads between drops contract and develop small end droplets (see the panel at  $t = 750$  ps) that subsequently move toward each other. Their collision yields a peak density (panel at  $t = 800$  ps). Not surprisingly, threads behave as the filaments described in Sec. III B. A similar pattern of alternating droplets and threads was observed in the study of the breakup of inviscid and irrotational capillary jets discussed in Ref. 38.

The lack of dissipation makes droplets and threads oscillate during the time elapsed by the simulation. Once formed, they execute a series of vibrations, being alternately compressed and elongated in the jet direction with an expected frequency of the order of  $\omega = \sqrt{8\gamma/m\rho_0 R_0^3}$ .<sup>39</sup> It has been pointed out that no obvious effects due to superfluidity have been observed on the breakup behavior of a liquid He jet.<sup>14</sup> Yet, Kolatzki *et al.*<sup>16</sup> have found that He droplets undergo shape oscillations that persist for much longer times than in the case of viscous drops, a signature of the superfluid character of these droplets.

We would like to mention that if only the cylinder density is perturbed and no axial velocity field is applied to it, we find that jet breaking proceeds as in Fig. 2, the only difference being that it takes more time for the instability to fully develop and eventually lead to jet fragmentation.

The actual time taken for the jet to break into droplets depends upon the amplitude of the initial density perturbation. It is defined as the time  $\tau_b$  it takes for the wave amplitude with the largest frequency to grow up to  $R_0$ ,<sup>21,40</sup>

$$R_0 = \delta_0 e^{\omega_{\max} \tau_b}, \quad (12)$$

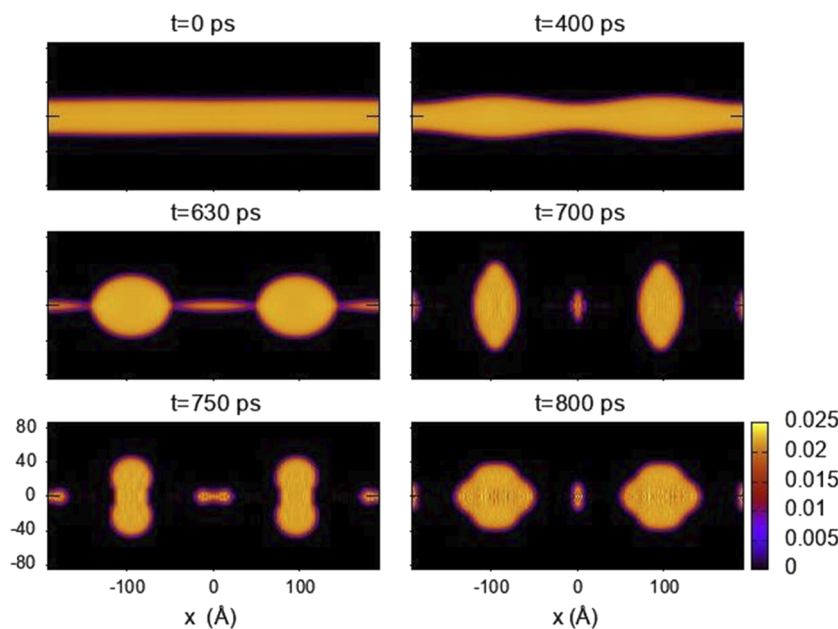
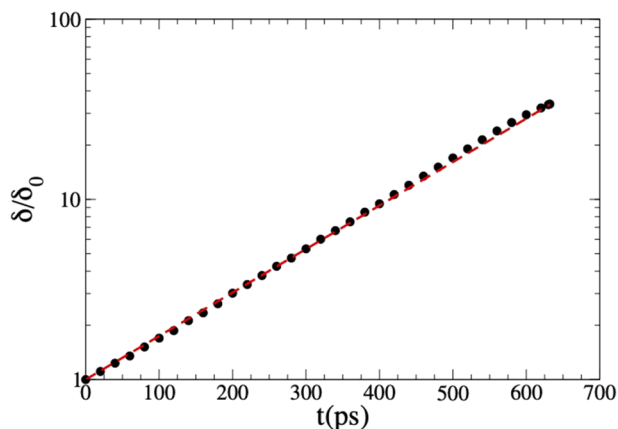


FIG. 2. Breaking dynamics of a cylinder subject to an axial perturbation of wavelength  $\lambda = 2\pi R_0/0.697$ . The color bar shows the atom density in units of  $\text{Å}^{-3}$ .



**FIG. 3.** Neck shrinking as a function of time, shown on a log-scale (see the text for the definitions of  $\delta$  and  $\delta_0$ ). The points are the numerical values obtained from the simulation, whereas the dashed line shows the prediction of linear theory.

where  $\omega_{\max}$  is given in Eq. (7). With our choice for the initial perturbation amplitude  $\delta_0$ , we have  $\tau_b = [\ln(R_0/\delta_0)]/\omega_{\max} = 695$  ps.

We have computed the dynamics of neck shrinking by monitoring during the real-time evolution the quantity  $\delta(t) = (R_{\max} - R_{\min})/2$ , where the radii  $R_{\max}$  and  $R_{\min}$  are measured at the two positions  $x = L/4$  and  $x = L/2$  (see Fig. 2). The calculated values for  $\delta(t)/\delta_0$  are shown in Fig. 3 on a logarithmic scale as a function of time and are compared with the quantity  $e^{\omega_{\max} t}$  predicted from linear theory. We find remarkable the good agreement, for the whole duration of the breaking process, between our simulations and linear theory.

Finally, we have also investigated another scenario when the jet is subject to a more general perturbation on the equilibrium density, i.e., we have started the real-time dynamics with the cylinder simultaneously perturbed by several axisymmetric perturbations of different wavelengths. In order to accommodate a reasonable number of modes with different wavelengths compatible with the PBC

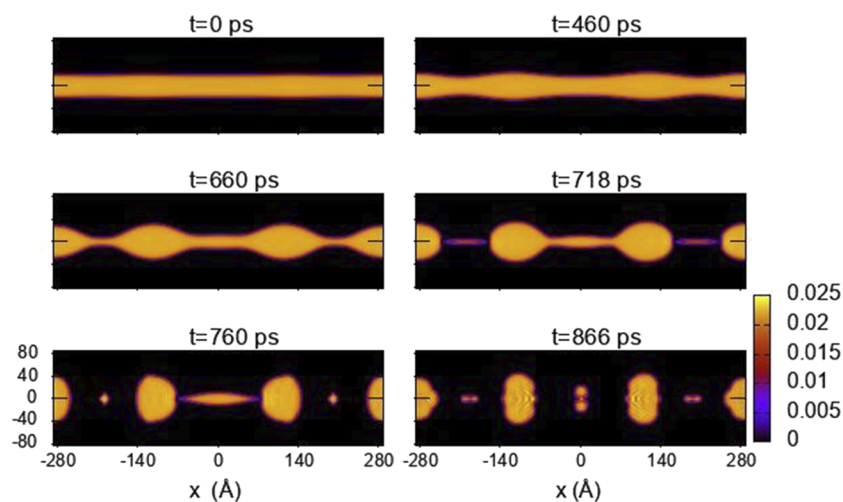
used here, we perform the simulation in a cell longer than the one shown in Fig. 2, with  $L$  equal to three times the critical wavelength associated with the fastest mode,  $\lambda_c = 2\pi R_0/0.697$ . We, therefore, consider an axially symmetric density perturbation given by a linear combination of six modes with small random amplitudes  $\varepsilon = \delta_0/R_0$  in the  $(-0.03, 0.03)$  range and wavelengths  $\lambda_c, 3\lambda_c, 3\lambda_c/2, \lambda_c/2, \lambda_c/4$ , and  $3\lambda_c/4$ , and perform a real-time simulation starting from such an initial state ( $t = 0$  panel in Fig. 4).

We show in Fig. 4 some snapshots of the density (on a plane containing the cylinder axis) taken during the real-time evolution of this system, where it appears that among the various modes, the one eventually dominating in the course of time is indeed the critical one, dictated by  $\lambda_c$ , which leads to the formation of three necks, eventually resulting in the fragmentation into three droplets. However, at variance with the case where the critical mode is the only one present (as shown in Fig. 2), the jet does not break up into equal-size droplets. For much longer filaments than the one investigated here, one might expect a distribution of slightly different drop sizes, some drops coming from the crests of the primary waves and others from the ligaments linking them. Determining the disturbance frequencies for jet breaking leading to the production of uniformly sized equidistant He drops has been one of the main concerns of a recent study<sup>16</sup> in view of their experimental use in, e.g., coherent diffraction imaging at x-ray free electron lasers.

We have thus seen that the He-DFT approach is able to address jet breaking and yield results in agreement with linear theory. This is a needed first step before carrying out the study of contracting <sup>4</sup>He filaments, which we address in the following.

## B. Contraction and fragmentation of free-standing filaments

Classical fluids jet breaking may lead not only to drops but also to filaments.<sup>17,18,38</sup> In the experiments, a filament is generated between two drops as a secondary product of breakup.<sup>41</sup> The appearance of filaments is obviously due to hydrodynamical instabilities developing in the jet but, to the best of our knowledge, the conditions under which they appear are not fully understood. Their



**FIG. 4.** Breaking dynamics of a cylinder subject to multiple wavelength axial perturbations as explained in the text. The color bar shows the atom density in units of  $\text{\AA}^{-3}$ .

study is further hampered by the difficulty of experimentally generating free cylindrical filaments whose properties could be compared with theoretical models describing their dynamical evolution. Only recently has it been possible to carry out a comprehensive experimental study of the breakup of normal liquid filaments based on an apparatus originally designed to replicate jet and droplet formation in an ink-jet printer, but on a larger scale to produce free cylindrical filaments.<sup>42</sup>

As with classical fluids, <sup>4</sup>He jet breaking may lead not only to droplets but also to filaments, as observed in experiments,<sup>14–16</sup> and we have shown that they may appear—not on purpose—between drops (see Figs. 2 and 4). Here we model these filaments as cylinders of radius  $R_0$  delimited by two hemispherical caps<sup>19</sup> and study, using the He-TDDFT approach, their contraction due to the effect of the surface tension for different values of the aspect ratio  $\Gamma = \bar{L}/R_0$ ,<sup>18</sup> where  $\bar{L}$  is the half-length of the filament from end-to-end. The configuration from which the real-time dynamics is initiated is a free-standing ideal filament (i.e., no density perturbation is applied), as is usually performed in numerical simulations of the contraction of viscous fluid filaments.<sup>19–21</sup>

We have investigated filaments with different values of the aspect ratio, namely,  $\Gamma = 4, 5, 6, 8, 10.5$ , and 15. Some of these values coincide with those studied in Ref. 19 at  $Oh = 0.001$ , which is considered to correspond to the inviscid regime. Our goal is to study how the initial aspect ratio  $\Gamma$  determines the fate of the filament, i.e., either contraction into a single liquid body (stable state) or breaking into two or more droplets. Experimentally,<sup>18</sup> it has been found for classical fluids that there is a critical initial aspect ratio  $\Gamma = 6 \pm 1$ , below which a liquid filament is stable irrespective of the  $Oh$  value, and above which the filaments tend to break into separate droplets.

In the following, we describe the most salient features found during the real-time evolution of superfluid <sup>4</sup>He filaments. All simulations discussed below are displayed as movies in the [supplementary material](#) accompanying this work. These movies last for longer times than those reported in the following figures. We

do not discuss the filament appearance for such long times because undamped excitations and especially the annihilation of vortex rings, as discussed in the following, tend to produce turbulence<sup>43,44</sup> whose description is beyond the scope of this paper.

### 1. Filament with $\Gamma = 4$

This is the shortest filament that we have investigated. Its time evolution is similar to that predicted for short filaments by classical calculations and experiments,<sup>18,19</sup> i.e., the filament contracts and oscillates back and forth without breaking. In the presence of some viscosity, the final configuration would be a single spherical droplet.

As shown by the temporal sequences in Fig. 5, a blood-cell shape develops in the transverse direction ( $y-z$  plane), which is clearly visible at  $t = 254$  ps, displaying an almost empty hole in the center (a toroidal shape; we recall that the system is axially symmetric around the  $x$  axis), before becoming compact again (frame at  $t = 300$  ps), and recovering a peanut-like shape along the filament axis (frame at  $t = 400$  ps). For longer times (not shown in the Figure but visible in the movies in the [supplementary material](#)), after extending transversally, it is drawn again into a compact droplet, originating a high density spot in its center. This high density spot relaxes and launches a series of density waves propagating inside the filament. This effect has been observed in previous simulations of the merging of two <sup>4</sup>He nanodroplets.<sup>43,44</sup>

### 2. Filament with $\Gamma = 5$

According to classical calculations and experiments, a filament with this value of  $\Gamma$  is also expected to display stable dynamics, oscillating back and forth without breaking.<sup>18,19</sup> Interestingly, simulation of this filament has disclosed the nucleation of quantized vortex rings, which appear in Fig. 6 as dark spots in the snapshots taken at  $t = 315$  ps and  $t = 380$  ps. For symmetry reasons, only pairs of quantized vortex-antivortex rings (vortex ring pairs with opposite circulation) can be nucleated. No such rings have been found in

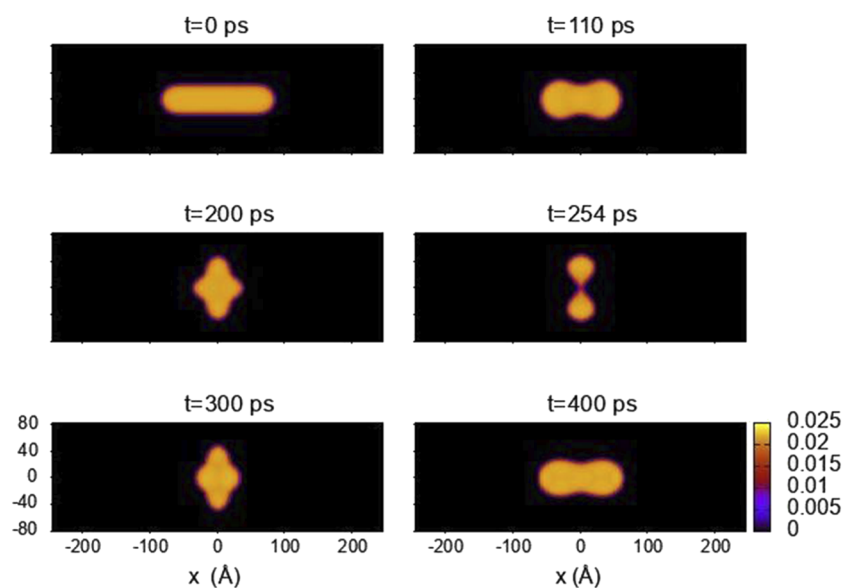
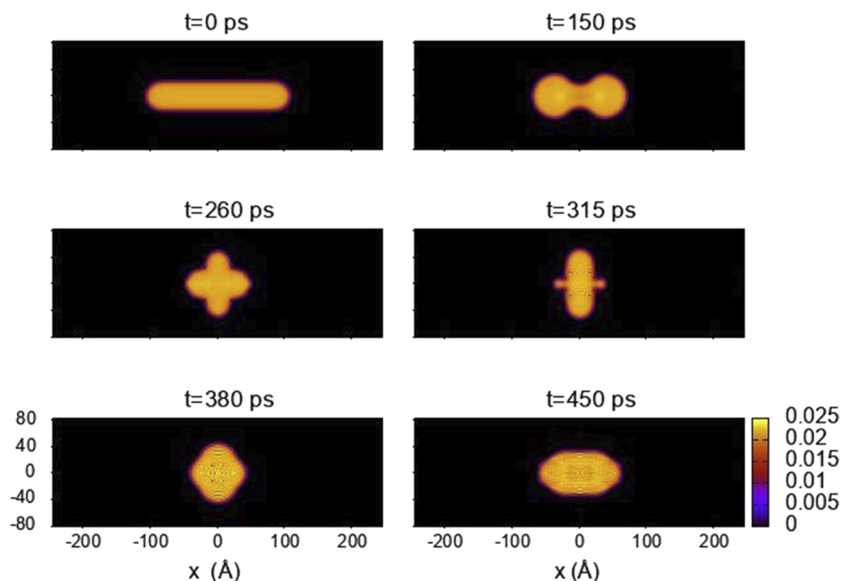


FIG. 5. Contraction of a filament with aspect ratio  $\Gamma = 4$ . The color bar shows the atom density in units of  $\text{\AA}^{-3}$ .



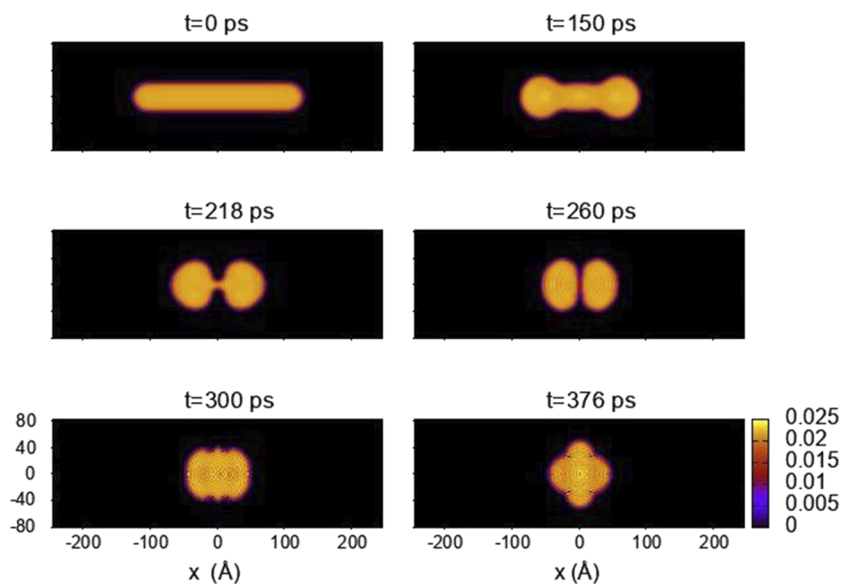
**FIG. 6.** Contraction of a filament with aspect ratio  $\Gamma = 5$ . The color bar shows the atom density in units of  $\text{\AA}^{-3}$ .

classical simulations carried out in the range of Ohnesorge numbers corresponding to the inviscid regime (below  $\sim 2 \times 10^{-3}$ ). Yet, Hoepffner and Paré have found classical vortex rings for Ohnesorge numbers in the  $0.002 < Oh < 0.1$  range but, surprisingly enough, not in the inviscid regime.<sup>21</sup> We will highlight the role of these vortices for the longer filaments discussed in the following, where they become effective in preventing the filament breaking.

In the case displayed in Fig. 6, vortices are nucleated during the contraction dynamics at surface indentations appearing between the end droplets or blobs and the rest of the cylindric filament (see the panel at  $t = 315$  ps); this requires some inertia, which can only be acquired when the aspect ratio of the filament is larger than a

critical value. Since this is the calculated filament with the smallest  $\Gamma$  value for which we see vortex rings nucleation, one should expect the appearance of vortex rings for filaments with  $\Gamma \geq 5$ . Once nucleated, vortices move to the bulk of the filament.

The filament end caps collapse (panel at  $t = 315$  ps) and launch additional vortex pairs in a small volume, which eventually annihilate, yielding an intense burst of density waves at later times (panel at  $t = 380$  ps). Eventually, the filament oscillates between the longitudinal and transverse directions, filled with density waves propagating inside the formed droplet (panel at  $t = 450$  ps). We find similarities with the  $L_0 = 5$  case in Ref. 19, but at variance with that reference, where



**FIG. 7.** Contraction of a filament with aspect ratio  $\Gamma = 6$ . The color bar shows the atom density in units of  $\text{\AA}^{-3}$ .

breakup appears by complex oscillations at  $t = 5.2 \tau_c$ , in our simulation the end caps are reabsorbed in the bulk of the resulting stable droplet.

### 3. Filament with $\Gamma = 6$

As shown in Fig. 7, the filament retracts and two end drops appear at the tips, clearly visible at around  $t = 150$  ps. Drops grow in size, and the filament between them contracts and shrinks into a thread (see, e.g., the configuration at  $t = 218$  ps). This thread collapses at  $t = 236$  ps, and the two droplets are temporarily apart, as shown in the panel for  $t = 260$  ps. However, due to the kinetic energy gained during the previous contraction stage, the two highly deformed fragments collide immediately after and merge again at  $t \sim 280$  ps to produce a single deformed droplet.

The collision of the fragments produces a high density spot at the contact region (between  $t = 262$  ps and  $t = 272$  ps; see movie in the [supplementary material](#)), which expands, yielding density waves propagating inside the filament (see the  $t = 300$  ps frame in Fig. 7). The merged drop presents surface indentations as those appearing, e.g., at  $t = 300$  ps. These indentations act as nucleation sites for quantized vortex rings, which remain close to the droplet surface. The cores of some of these vortices are clearly visible in the frame at  $t = 376$  ps. Notice that these vortices do not contribute to the escape from end-pinching since the thread connecting the end drops has collapsed before. The density is no longer smooth; rather, it is strongly perturbed by the presence of density waves produced by the merging of the two fragments.

The evolution of this filament is similar to the  $L_0 = 6.0$  filament shown in Fig. 5 of Ref. 19. For superfluid  $^4\text{He}$ , we have found that the filament temporarily breaks into two deformed drops at  $t = 3.8 \tau_c$ , similar to the value one can read in Fig. 5 of that reference. However, in our case, the drops collide and merge again, whereas in Ref. 19, they seem to remain separated. Another difference between classical and superfluid filaments is the appearance of quantized vortex rings and their subsequent annihilation.

### 4. Filament with $\Gamma = 8$

The dynamical evolution of this filament is shown in Fig. 8. As for the previous case, end drops developed and were clearly visible already after  $t \sim 100$  ps. The main filament connecting the end drops shrinks, and a thin neck develops at the drop-filament contact region, which starts pinching off the filament with two necks that reach their smallest radius at  $t = 254$  ps. Before they completely shrink, vortex rings nucleate close to the necks at about  $t = 258$  ps and are clearly formed at  $t = 270$  ps. The streamlines of the superflow are drawn in the top panel of Fig. 9 for the configuration at  $t = 290$  ps, clearly showing the characteristic pattern of lines wrapping the vortex core positions.

These vortex rings prevent necks from pinching, as they reopen immediately after their appearance (see, e.g., the frame at  $t = 290$  ps), similarly to the mechanism discussed by Hoepffner and Paré.<sup>21</sup> A flow through the neck develops because of the retraction and, according to these authors, this flow may detach into the jet downstream of the neck when fluid viscosity exceeds a threshold ( $Oh \gtrsim 2 \times 10^{-3}$ );<sup>21</sup> this sudden detachment creates a vortex ring that strongly modifies the flow pressure, and fluid is transported back into the neck, which in turn reopens. It is remarkable that the same happens in the case of superfluid  $^4\text{He}$  despite the lack of viscosity. At  $t = 330$  ps, another pair of vortex rings is nucleated at the droplet-filament indentation, preventing pinching again. Finally, vortex-antivortex rings annihilate and disappear from the system, producing a burst of density waves as a result.

The movie in the [supplementary material](#) shows the appearance of surface protrusions at  $t = 452$  ps, which act as vortex nucleation sites, and their collapse yields a high density spot. Eventually, the contracted filament is permeated by a large number of vortex rings at  $t = 488$  ps. This is at variance with the classical, inviscid fluid description.

The evolution of this filament can be compared to that corresponding to  $L_0 = 8.0$  shown in Fig. 5 of Ref. 19. Besides the vortex

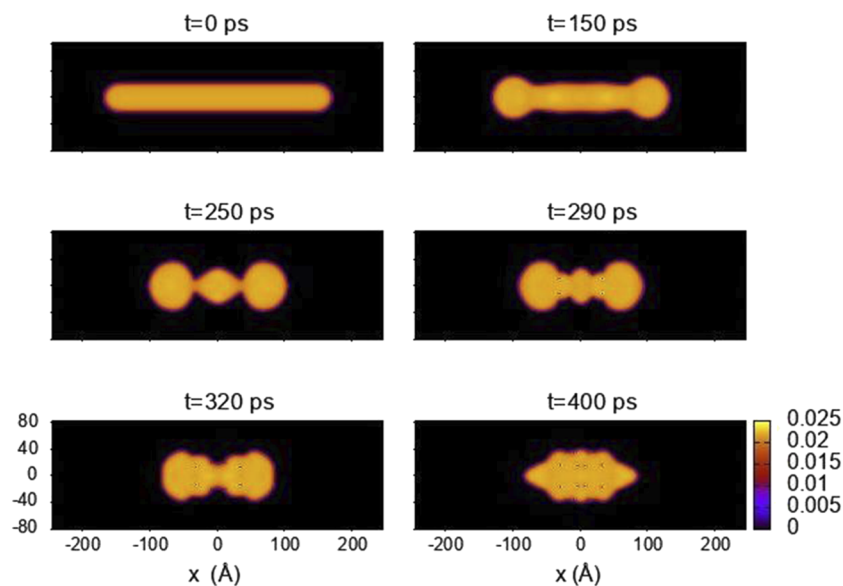
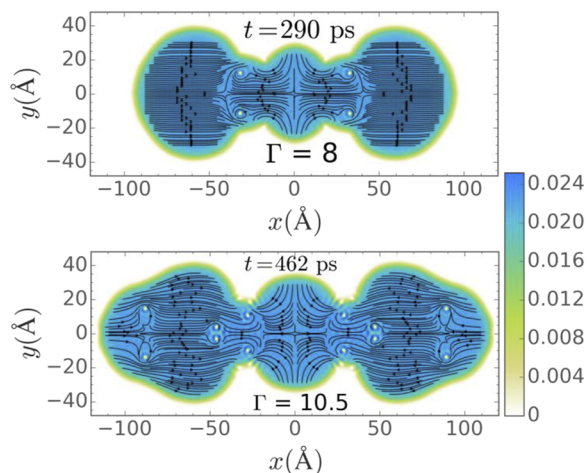


FIG. 8. Contraction of a filament with aspect ratio  $\Gamma = 8$ . The color bar shows the atom density in units of  $\text{\AA}^{-3}$ .



**FIG. 9.** Superfluid streamlines corresponding to the configurations  $\Gamma = 8$  at  $t = 290$  ps (top) and  $\Gamma = 10.5$  at  $t = 462$  ps (bottom). The color bar shows the atom density in units of  $\text{\AA}^{-3}$ .

rings phenomenology, which is absent in the simulations of that reference, in our case end-pinching (i.e., the detachment of two droplets from the two ends of a retracting filament) strictly never happens. The  $^4\text{He}$  filament gets the closest to it at  $t = 4.1 \tau_c$ , whereas the time for the filament breakup by end-pinching read from Fig. 5 of Ref. 19 is  $t \sim 4.6 \tau_c$ .

### 5. Filament with $\Gamma = 10.5$

Similarly to the previous cases, end drops develop as shown in Fig. 10. A more violent approach is expected because the filament is longer and end drops have more time to accelerate under the traction exerted by surface tension. The filament connecting the

end drops contracts and necks appear at the drop-filament contact region, as shown at  $t = 250$  ps, which start pinching. The neck shrinks to a minimum at  $t = 256$  ps, escaping from end-pinching again because vortex rings are nucleated at  $t \sim 260$  ps.

Vortex rings detach from the neck and move toward the bulk of the end drops (frame at  $t = 300$  ps). The remaining filament develops bulges, which evolve into a more complex structure (frame at  $t = 400$  ps).

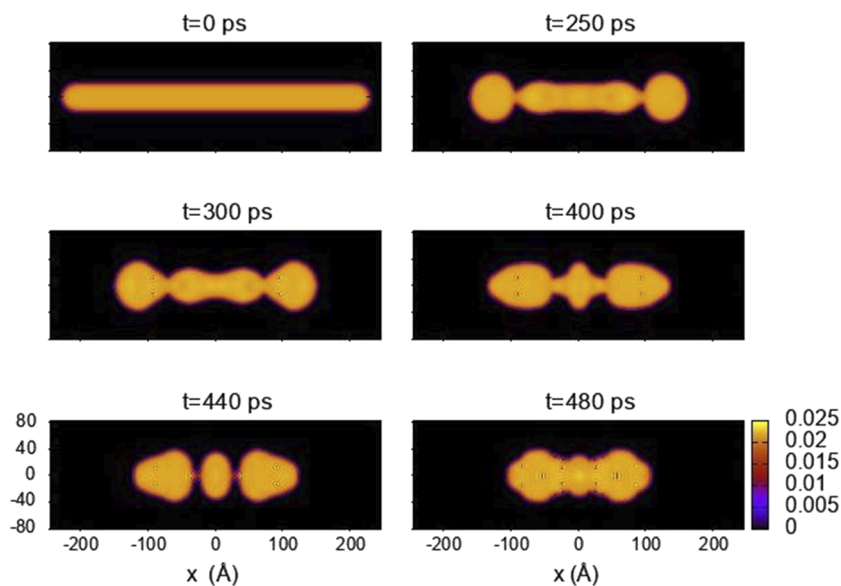
The snapshot at  $t = 440$  ps shows an almost complete fragmentation. However, due to the opposite velocities acquired during the early stages of the contraction, the three fragments merge again. Other vortex rings are created in the process and nucleated at the necks during the re-merging, as shown in the frame at  $t = 480$  ps. The streamlines of the superflow are drawn in the bottom panel of Fig. 9 for the configuration at  $t = 462$  ps.

Vortex ring annihilation at later times (see movie in the supplementary material) produces density waves arising from the collapse of their cores. This is a phenomenon that we have not observed in the merging of  $^4\text{He}$  droplets<sup>43,44</sup> or the shrinking of a vortex ring before it collapses. It is interesting to see that these small vortex rings travel toward the tips of the filament, evaporating from them. Eventually, vortex rings disappear, and the contracted filament enters a complex dynamic regime, hosting plenty of density waves until the end of the simulation.

The evolution of this filament should be similar to that corresponding to  $L_0 = 10.0$  shown in Fig. 5 of Ref. 19. Besides the vortex rings phenomenology and wave dynamics, in our case, end-pinching strictly never occurs. The filament gets close to it at  $t = 4.1 \tau_c$  (254 ps) and especially at  $t = 7.0 \tau_c$  (432 ps), whereas the breakup time by end pinching read from Fig. 5 of Ref. 19 is  $t \sim 4.8 \tau_c$ .

### 6. Filament with $\Gamma = 15$

This is the largest filament we have investigated. In classical simulations of sufficiently long filaments and small  $Oh$  numbers, as the filament contracts, it will succumb to end pinching<sup>18,20,45</sup>



**FIG. 10.** Contraction of a filament with aspect ratio  $\Gamma = 10.5$ . The color bar shows the atom density in units of  $\text{\AA}^{-3}$ .

even in cases where the Rayleigh–Plateau instability is expected to develop, subsequently resulting in the filament breaking up into several drops. However, this instability does not occur, suggesting that the timescale for the Rayleigh–Plateau instability to grow is much larger than the timescale for the filament to fully contract, even for long filaments. The same happens in the case of  $^4\text{He}$ , as can be seen from Fig. 11.

In the case of superfluid  $^4\text{He}$ , however, end-pinching is again hindered by vortex ring generation. The temporal sequence is similar to the  $\Gamma = 8$  and  $\Gamma = 10.5$  cases, except that the number of necks has increased. Well developed end drops appear at  $t = 100$  ps, with well developed necks at  $t = 160$  ps. Figure 11 shows that the end drops nearly pinch-off at  $t = 248$  ps, but at  $t = 264$  ps, one may see vortex rings appearing at the necks, hindering end-pinching. The vortex rings detach from the neck and move toward the bulk of the end drops, and bulges appear in the filament close to the end drops (panel at  $t = 290$  ps). Bulges evolve to bulbs and, similarly to the  $\Gamma = 8$  and  $\Gamma = 10.5$  cases, intermediate drops develop during the time evolution whose number increases with the length of the filament, as also observed in the simulations of classical low viscosity ( $0.003 \leq Oh \leq 0.02$ ) filaments.<sup>41</sup>

The evolution of this filament should be compared to that corresponding to  $L_0 = 15.0$  shown in Fig. 5 of Ref. 19. Besides the phenomenology of vortex rings proliferation, in this case, end-pinching never occurs. End drops are close to detach at  $t = 4.02 \tau_c$  (247 ps) but escape pinch off because of vortex ring nucleation, whereas the filament breakup time read from Fig. 5 of that reference is  $t \sim 4.8 \tau_c$ .

Finally, we have computed the contraction velocity for all the investigated filaments. We have defined the position of the tip of the filament as the location of its sharp surface (that at which the density equals  $\rho_0/2$ ) on the  $x$  axis.

Figure 12 shows the displacement of the tip position as a function of time for the studied filaments. It appears that all curves collapse onto the same curve up to  $t \sim 170$  ps ( $2.76 \tau_c$ ). Consequently, within this range of time, the retracting velocity is independent from the aspect ratio  $\Gamma$ . For times in the  $50 \text{ ps} \leq t \leq 170 \text{ ps}$  range, all filaments accurately follow the line with the slope equal to the Taylor–Culick velocity  $v = R_0/\tau_c = 0.348 \text{ \AA/ps}$ , which is the

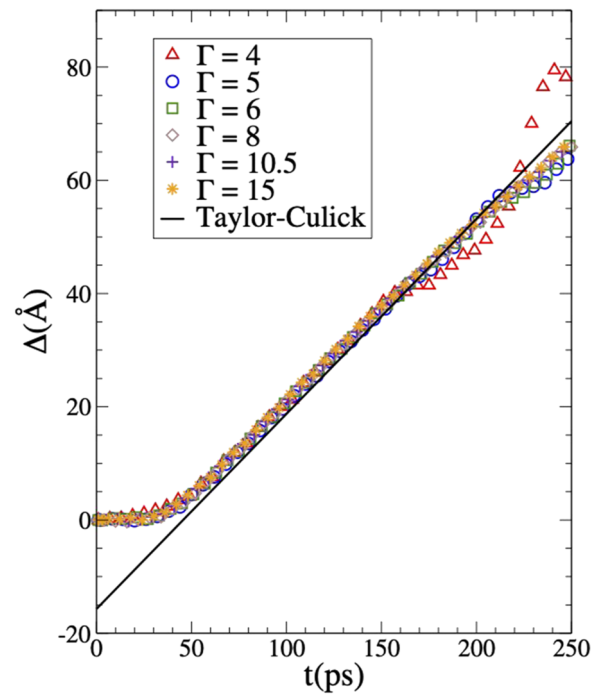


FIG. 12. Contraction of filaments with different aspect ratios as a function of time. The slope of the solid line is the theoretical contraction velocity  $R_0/\tau_c$ .

relevant velocity scale expected for the retraction process and was originally proposed<sup>46,47</sup> as the steady-state velocity of a capillary-driven retracting inviscid planar liquid where inertia effects balance the capillary forces acting on the system. For longer times, the behavior changes because there are either filament oscillations, changes in the tip shape, or both. The shorter the filament, the earlier these deviations start to show up. The retracting velocity of liquid filaments has been studied for Ohnesorge numbers  $Oh \geq 0.1$ ,<sup>48</sup> finding

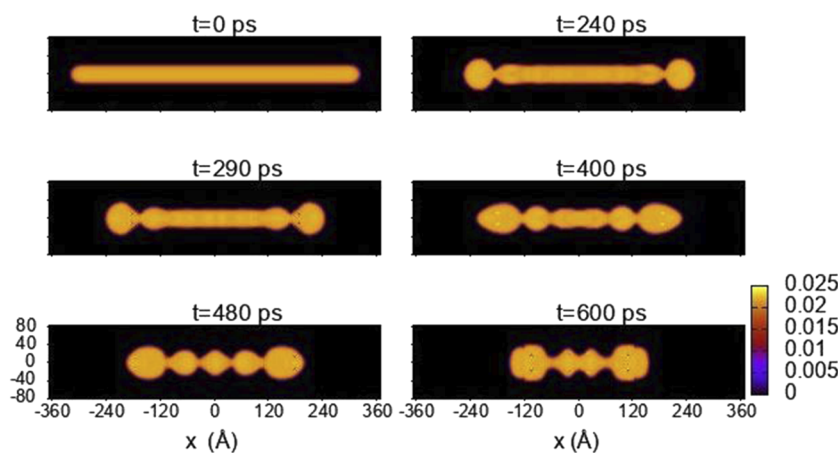


FIG. 11. Contraction of a filament with aspect ratio  $\Gamma = 15$ . The color bar shows the atom density in units of  $\text{\AA}^{-3}$ .

that the tip dynamics are characterized by an oscillating velocity whose mean value is close to the Taylor–Culick prediction. These oscillations have also been found for  $Oh = 0.05$  in the  $\Gamma = 20$  case.<sup>48</sup> In superfluid  $^4\text{He}$ , though, we do not observe any oscillation in time of the tip retraction velocity.

#### IV. SUMMARY AND CONCLUDING REMARKS

We have studied the instability and breakup of nanoscopic superfluid  $^4\text{He}$  jets and filaments within He-DFT at zero temperature. We find that the fragmentation of long cylindrical jets closely follows the predictions of the linear theory for inviscid fluids, resulting in the formation of larger droplets intercalated with smaller satellite droplets.

While some of our results for the contraction of free-standing filaments are consistent with those obtained in the inviscid regime, which correspond to Ohnesorge numbers smaller than  $2 \times 10^{-3}$ ,<sup>19</sup> the novelty with respect to previous calculations for classical inviscid filaments is the appearance of quantized vortex rings in filaments with aspect ratio  $\Gamma > 5$ .

Non-quantized vortex ring nucleation in the region connecting the end drops with the rest of the filament plays a central role in escaping filament breakup in the low-to-intermediate viscosity regime characterized by Ohnesorge numbers in the  $0.002 < Oh < 0.1$  range.<sup>21</sup> Our simulations show that a similar mechanism, associated with quantized vortex rings, is active in the superfluid regime at zero temperature, mostly preventing droplet formation through end-pinching. Vortices are also nucleated at surface protrusions appearing in the course of filament oscillations, similar to those found in the merging of  $^4\text{He}$  droplets. As a result, filaments are permeated by vortex-antivortex ring pairs, whose annihilation yields phonon/roton bursts that may leave the filament in a turbulent state.<sup>43,44</sup>

A key question is why vortex rings, which have appeared in the solution of the Navier–Stokes equation in the  $0.002 < Oh < 0.1$  regime, cease to appear in the inviscid regime<sup>19,21</sup> whereas we have found them in the superfluid regime within the He-DFT approach. It is known that the Gross–Pitaevskii and He-TDDFT equations, appropriate for superfluids, do not reduce to the zero-viscosity limit of the Navier–Stokes equation (Euler equation) for a barotropic fluid in irrotational flow.<sup>27</sup> In the superfluid case, an extra term appears involving the gradient of the expression,

$$Q = \frac{\hbar^2}{2m} \frac{\nabla^2 \rho^{1/2}}{\rho^{1/2}}, \quad (13)$$

the so-called quantum pressure term. This term, which is missing in any classical approach, plays an important role when the density is highly inhomogeneous, as it happens near the core of a quantized vortex. At variance, it is an ingredient naturally included in the Schrödinger He-TDDFT Eq. (3) and time-dependent Gross–Pitaevskii equation as well.<sup>27</sup>

Notice that the vortices found in this work are vortex rings. These vortices carry no angular momentum and are created in vortex–anti-vortex ring pairs. We have found in the simulations that either they annihilate upon vortex–antivortex ring collisions, yielding an intense roton burst, or “evaporate” from the filament surface, leaving in both cases vortex-free droplets. The commonly accepted

basic mechanisms for vortex nucleation in  $^4\text{He}$  droplets are friction against the nozzle when exiting droplets are still in the normal phase or by droplets collisions within the droplet beam at some distance from the nozzle. The capture of impurities by  $^4\text{He}$  droplets may also yield vortex rings and vortex loops nucleation, but they are rather short-lived.<sup>49,50</sup> It is worth noticing the interest in knowing whether droplets formed upon breakup of the  $^4\text{He}$  jet carry or not vortices, as their presence affects how nanostructures are assembled within  $^4\text{He}$  droplets.<sup>51</sup>

We have shown that the He-DFT approach, which is a suitable method to describe pure and doped superfluid  $^4\text{He}$  nanodroplets, can also address superfluid  $^4\text{He}$  jet breaking and the contraction of superfluid  $^4\text{He}$  filaments. The simulations have been carried out for nanoscopic structures with an  $R = 21.5 \text{ \AA}$  radius. The experimental observations, however, have been made on systems 2–3 orders of magnitude bigger, so it is natural to wonder if our results could be related to any experimental observation. We want to point out that we have carried out simulations for the  $\Gamma = 15$  filament using a radius twice as larger and have sensibly found the same results. The crucial point is to simulate structures with a radius large enough so that they have developed a bulk region clearly distinguishable from the surface region where the density goes smoothly to zero (finite surface width), as it happens for the  $R$  value we have chosen (see Fig. 1). Once this condition is fulfilled, the results should reflect the behavior of larger, mesoscopic systems, and the parameter to scale the simulations to larger sizes is, as for classical systems, the aspect ratio  $\Gamma$ . The situation is very similar to that of rotating  $^4\text{He}$  droplets, where the experiments have been carried out for droplets several orders of magnitude bigger than the simulations<sup>7,8</sup> and, despite this, the comparison with the simulations is meaningful.<sup>52</sup> There too, the drops were large enough to display distinct bulk and surface regions.

Yet, our simulations have some unavoidable limitations that prevent a detailed comparison with experiments. On the one hand, our approach is strictly a zero temperature approach, and there is no dissipation; energy can only be lost by atom evaporation. Due to the limited time elapsed by the simulation, at the end of the simulations, droplets and filaments are still in an excited state where the system is permeated by density waves that should be damped in the long term by any residual viscosity remaining in the system or by atom evaporation. On the other hand, we have found that upon filament breaking, the resulting fragments have a tendency to merge again into a single droplet. Two effects combine to favor this behavior. First, fragments, which are nanoscopic, have a non-zero surface width that helps recombination due to the overlap of the densities tails. Second, the contraction velocity acquired by the filament in the early stages of the contraction tends to push the two highly deformed drops together even if they are temporarily apart. One should also consider the role of long-range van der Waals attractive interaction between separated fragments, which may also contribute to their merging. For the much larger sizes in the experiments, however, the vdW forces are expected to be negligible. In fact, the force between two spherical particles of diameter  $D$  made of  $q$  atoms per unit volume interacting via the two-body vdW interaction  $\lambda/r^6$  is<sup>53</sup>  $F \propto -\tilde{F}(x)/D$ , where  $x = d/D$ ,  $d$  being the distance of closest approach between the spheres surfaces, and  $\tilde{F}(x) \sim -1/(24x^2)$  ( $x \ll 1$ ). Therefore, for the sizes encountered in experiments, the vdW attraction between fragments will be much reduced,

if not negligible, meaning that once a filament breaks into two fragments, recombination into a single droplet due to the vdW attraction is unlikely.

## SUPPLEMENTARY MATERIAL

See the [supplementary material](#) for the video files showing the real time evolution of the processes discussed in the present work.

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## AUTHOR DECLARATIONS

### Conflict of Interest

The authors have no conflicts to disclose.

### Author Contributions

F.A., M.B., and M.P. contributed equally to this work.

**Francesco Ancilotto:** Conceptualization (equal); Data curation (equal); Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Methodology (equal); Project administration (equal); Resources (equal); Software (equal); Supervision (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Manuel Barranco:** Conceptualization (equal); Data curation (equal); Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Methodology (equal); Project administration (equal); Resources (equal); Software (equal); Supervision (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Martí Pi:** Conceptualization (equal); Data curation (equal); Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Methodology (equal); Project administration (equal); Resources (equal); Software (equal); Supervision (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal).

## DATA AVAILABILITY

The data that support the findings of this study are available within the article and its [supplementary material](#).

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