Density of Superfluid Helium Droplets

Jan Harms, J. Peter Toennies

Max-Planck-Institut für Strömungsforschung, 37073 Göttingen, Germany

Franco Dalfovo

Dipartimento di Fisica, Università di Trento, 38050 Povo, Italy

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Abstract

The classical integral cross sections of large superfluid $^4\text{He}_N$ droplets and the number of atoms in the droplets ($N = 10^3 - 10^4$) have been measured in molecular beam scattering experiments. These measurements are found to be in good agreement with the cross sections predicted from density functional calculations of the radial density distributions with a 10–90% surface thickness of 5.7Å. By using a simple model for the density profile of the droplets a thickness of about 6–8Å is extracted directly from the data.
I. INTRODUCTION

The study of the properties of $^4\text{He}$ clusters is currently an active area of theoretical and experimental research \([1,2]\). The interest is based on the fact that $^4\text{He}$ clusters provide an easily accessible example of a finite size quantum system of strongly correlated particles. One of the primary aims of He cluster research is to search for manifestations of superfluidity in mesoscopic systems. Recent spectroscopic studies of glyoxal molecules embedded in $^4\text{He}$ droplets with about 5000 atoms produced in free jet expansions provide the first evidence that these droplets are indeed superfluid \([3]\). The internal temperature of these droplets has also been measured spectroscopically to be about 0.4 $K$ \([4]\) in agreement with theory \([5]\).

Even though there has been notable progress in theoretical work on He clusters during the last years the agreement between theory and experiments is not yet entirely satisfactory \([1]\). The implementation of *ab initio* calculations still remains difficult for inhomogeneous systems with more than a few hundred atoms, while phenomenological theories, which work well for describing macroscopic properties, are often not adequate for systems of finite size. A major progress to fill this gap has been made in the development and use of density functional (DF) theory \([6]\).

The understanding of the density profiles in the surface region of a quantum fluid has long been considered a very important basic problem \([7]\). For this reason significant effort has gone into the experimental determination of the thickness of the bulk liquid-vapor $^4\text{He}$ interface \([7]\). At least three different experimental methods have been used to measure the surface thickness of bulk liquid $\text{He}$. The first involved atomic scattering experiments \([8]\) yielding an interfacial width (10–90%) of about 4Å. A surface thickness of about 9Å was measured with an ellipsometric method \([9]\). Finally, x-ray measurements give a surface thickness of the order of 8Å, providing also information on the shape of the interface \([10]\). In droplets the surface atoms represent a significant fraction of the total number of atoms. For example, in a droplet with 5000 atoms about one half of the atoms are located in the surface region. Thus the physical properties of $^4\text{He}$ droplets are strongly influenced by the
surface. Moreover, the surface region is particularly interesting because recent calculations predict that the Bose-condensed fraction, which in bulk $^4\text{He}$ is about 10%, approaches 100% in regions where the density is still within 10% of the bulk density [1].

In this paper the liquid-vapor interface of large $^4\text{He}$ droplets ($N = 10^3 - 10^4$) is investigated by a new method. The experiment consists in measuring both the integral cross section of the droplets and also their number size distributions. From the integral cross section and the average number of atoms an effective droplet volume as well as an average density is determined. The results are compared with theoretical predictions. Although a large number of calculations have been performed for the density profiles of small $^4\text{He}$ clusters ($N < 10^3$) [1,12–18] no calculations for the large droplets investigated here have been reported so far. In the present work DF theory is used to calculate the density profiles of large $^4\text{He}$ droplets. The density functional used was recently introduced by the Orsay-Trento collaboration [3]. This functional provides density profiles and energies [12] very close to the results of ab initio Monte Carlo calculations [13-15] in the case of small clusters and has the advantage that it can easily be applied to droplets with several thousands of atoms. These density profiles provide for a direct comparison with the experimental data and good agreement is found within the experimental errors.

The droplet beam scattering apparatus is described in the following section. The procedure used to measure the number of atoms in the droplets and their classical integral cross sections are described in Section II A and II B. In Section II C the experimental results are presented and in Section II they are compared with the DF predictions. A final discussion is given in Section IV.

**II. EXPERIMENTAL**

Since the experimental methods and the apparatus used in this work have already been described in some detail in previous publications [19,20] the experimental procedure will be only briefly described here. The $^4\text{He}$ droplet beam is produced by a free jet expansion of
He from a stagnation source chamber at a high pressure and at low temperature (typically $P_0 = 5 - 100 \text{ bar}, T_0 = 4 - 30 \text{ K}$) through a thin walled ($20 \mu m$) nozzle with $5 \pm 1 \mu m$ diameter. After the droplets have passed the skimmer and several differential pumping stages they are scattered by a secondary beam produced in another free jet expansion beam source which crosses the droplet beam at an angle of $40^\circ$. The scattered droplets are detected with a homemade electron impact ionizer optimized for a high ionization efficiency followed by a magnetic mass spectrometer. The detector can be rotated around the scattering region in the plane of the two beams. To avoid capture collisions with the residual gas the background non-helium pressures in the differential pumping stages between source and scattering chamber were kept below $10^{-7} \text{ mbar}$.

A. The Number of Atoms in the Droplets

The atom number distribution $P(N)$ of the helium droplets is determined from the angular distribution resulting from scattering by the secondary beam. A small portion (5–10%) of the droplets is deflected by the momentum transfer imparted by single collisions of the secondary beam gas atoms, most of which are captured by the droplets \[20\]. The measurements described in the present study have been carried out at source pressures of $P_0 = 40 \text{ bar}$ and $P_0 = 80 \text{ bar}$ and nozzle temperatures between $T_0 = 13 \text{ K}$ and $T_0 = 26 \text{ K}$. For these conditions the droplet velocity distributions are very narrow ($\Delta v/v \approx 2\%$) with well known mean speeds \[21\]. The secondary beam gases Ar and Kr were expanded from stagnation temperatures of about $300 \text{ K}$ and several hundred millibar of stagnation pressure from a $40 \mu m$ diameter nozzle. The secondary beam contains no appreciable amount of dimers or larger clusters, and has a narrow velocity distribution ($\Delta v/v \approx 20\%$).

The angular distribution of the deflected droplet beam is measured by rotating the detector in small angular increments (typically $300 \mu rad$) around the scattering center. Due to the large mass of the droplets the deflection angles are very small and a high angular resolution is necessary. This is achieved by collimating the incident beam with a $50 \mu m$ slit.
in front of the scattering center and a 25 \( \mu m \) slit in front of the detector. The width of the expansion zone and the broadening due to collisions with the residual background gas (mostly helium) leads to a FWHM of the beam profile of about 1.5 mrad which defines the effective angular resolution.

Fig. 1 shows three typical measured angular distributions with and without a flag in front of the secondary beam [19]. The small difference between the two signals is attributed to droplets which were deflected after capturing a secondary beam atom. Assuming complete momentum transfer the angle of deflection is directly dependent on the number of atoms in droplet. As is discussed in more detail below, the present experiments give further justification for the hypothesis of complete momentum transfer. The measured droplet atom number distributions can be very well fitted with a log-normal-distribution [19,20]:

\[
P(N) = \frac{1}{\sqrt{2\pi N\delta}} \exp\left[\frac{-(\ln N - \mu)^2}{2\delta^2}\right],
\]

where the mean number of atoms \( \overline{N} \) and the width (FWHM) are

\[
\overline{N} = \exp\left(\mu + \frac{\delta^2}{2}\right)
\]

and

\[
\Delta N_{1/2} = \exp\left(\mu - \delta^2 + \delta\sqrt{2\ln 2}\right) - \exp\left(\mu - \delta^2 - \delta\sqrt{2\ln 2}\right),
\]

respectively. The FWHM is comparable with \( \overline{N} \). Table I lists the results for \( \overline{N} \), \( \Delta N_{1/2} \), \( \delta \) and \( \mu \), measured using Ar and Kr as deflecting atoms for a wide range of different \( ^4 \)He source pressures and temperatures. The values of \( \overline{N} \) and \( \Delta N_{1/2} \) obtained using Ar or Kr for identical \( ^4 \)He source conditions do not always agree exactly, since they depend sensitively on the nozzle-diameter and on the nozzle-skimmer distance which was optimized for each experiment and differed slightly. In particular the mean droplet sizes for \( P_0 = 80 \text{ bar} \) with krypton as secondary gas are somewhat smaller by about 30\% since the nozzle used in these experiments had a diameter of only about 4 \( \mu m \) as estimated from the total gas flow into the nozzle chamber. These deviations however have no appreciable effect on the
average densities. The latter depend on the number of atoms in the droplets and the integral cross sections, both of which are determined in the same experiment under identical source conditions (see below).

B. Integral Cross Sections

The integral cross sections of the droplets are determined by measuring the attenuation of the forward peak in the deflection pattern (see Fig. 1). The attenuation is related to the integral cross section $\sigma$ of the droplets according to Beer’s law [20],

$$\frac{I}{I_0} = \exp \left[ -\frac{\sigma n_{\text{sec}} L_{\text{eff}} v_{\text{rel}} F_{a0}}{v_{\text{drop}}} \right],$$  \hspace{1cm} (4)

where $I$ and $I_0$ are the intensities of the $^4\text{He}$ droplet beam with and without attenuation, $n_{\text{sec}}$ is the number density of the secondary beam gas atoms in the scattering center and $L_{\text{eff}}$ is the effective length of the scattering volume. Moreover, $v_{\text{drop}}$ is the speed of the droplets and $v_{\text{rel}}$ the relative collision velocity. $F_{a0}$ takes account of the smearing resulting from the velocity distributions of the two nozzle beams [22] and leads to a correction smaller than about 1%.

The product of the density in the scattering center and the effective length of the scattering volume ($n_{\text{sec}} \cdot L_{\text{eff}}$) was calibrated to within an error of approximately 5% by measuring the attenuation of a nearly monoenergetic helium atomic beam, for which the integral cross section can be accurately calculated quantum mechanically from the well established interaction potential [23]. The density was calibrated three times with krypton and two times with argon at different collision energies and there was no evidence of systematic errors. The values of the absolute integral cross sections of the droplets are also listed in Table I.

The interpretation of the integral cross sections in Eq. (4) is straightforward only in the ideal case of an infinite angular resolution. In this case one gets the quantum mechanical integral cross section, which includes the forward peaked diffraction part. In the present case of finite angular resolution the measured integral cross section is smaller since a fraction of the
forward diffraction part of the differential cross section is not included. In order to estimate this correction, the quantum mechanical differential cross section for elastic scattering was calculated using a partial wave expansion method [24]. The calculations, which are described in detail in Appendix A, indicate that the contribution of the diffraction part to the elastic cross section is negligible with the angular resolution of our apparatus. Thus the measured integral cross section $\sigma_{\text{int}}^{\text{app}}$ turns out to be very close to the classical cross section $\sigma_{\text{class}}$. The two cross sections agree to within 1% for droplets with 1000 atoms and for larger droplets the agreement is even better.

The finite width of the beam-profile also has the effect that the deflections of the largest droplets in the atom number distributions are too small to lead to a measurable attenuation of the beam. This error however can be estimated since the atom number distributions of the droplets and the beam-profiles $S(\Theta)$ are both well known from the measurements. The calculation yields a correction of about 1% for mean atom numbers of $10^3$ and of about 10% for droplets with $10^4$ atoms.

C. Mean Droplet Densities

Measured values of the integral cross section, $\overline{\sigma}$, as a function of the mean atom numbers $\overline{N}$ are reported in Fig. 2. The values obtained with argon and krypton as secondary gas fall on a common curve and so we can conclude that the integral cross section is independent of the nature of the secondary beam, within the estimated accuracy. The overall experimental errors are estimated to be about 5% and result mainly from the uncertainty in the determination of the density of the secondary beam atoms in the scattering volume.

From the measured cross section an effective mean density $\overline{\rho}$, defined as the density of a uniform sphere with a sharp step edge (liquid drop model) having the same classical integral cross section, is determined

$$\overline{\rho} = \frac{3}{4} \sqrt{\frac{\pi}{\overline{\sigma}}} \overline{N}.$$  (5)
The values of $\bar{\rho}$, normalized to the bulk helium density $\rho_{\text{bulk}} = 0.0218$ Å$^{-3}$, are given in the last column of Table I and the integral cross sections for spheres of different density are shown in Fig. 2 as dashed lines. Here the average integral cross section $\bar{\sigma}$ is calculated from the measured log-normal atom number distributions $P(N)$ by means of the following equation:

$$\bar{\sigma}(N) = \int_0^\infty P(N) \pi^\frac{1}{3} \left( \frac{3N}{4\rho} \right)^{\frac{2}{3}} dN . \quad (6)$$

As can be seen, the experimental effective droplet density goes from about $0.5 \cdot \rho_{\text{bulk}}$, for droplets with $10^3$ atoms, to $0.7 \cdot \rho_{\text{bulk}}$ for droplets with $10^4$ atoms. This trend is consistent with the fact that a significant part of the atoms are in the outer surface region, where the density is less than the bulk density.

### III. COMPARISON WITH THEORY

For comparison with the above experimental results the integral cross sections were calculated using density functional theory. These calculations were carried out with the improved density functional recently developed [3] for the accurate calculation of $^4$He droplet properties. The energy of the system is assumed to be a functional of the complex function $\Psi$:

$$E = \int dr \, \mathcal{H}[\Psi, \Psi^*] , \quad (7)$$

$$\Psi(r, t) = \Phi(r, t) \exp \left( \frac{i}{\hbar} S(r, t) \right) . \quad (8)$$

The real function $\Phi$ is related to the diagonal one-body density by $\rho = \Phi^2$, while the phase $S$ fixes the velocity through the relation $\mathbf{v} = (1/m) \nabla S$. In the calculation of the ground state, only states with zero velocity are considered, so that the energy is simply a functional of the one body density $\rho(r)$. A natural representation is given by

$$E = \int dr \, \mathcal{H}[\rho] = E_c[\rho] + \int dr \frac{\hbar^2}{2m} (\nabla \sqrt{\rho})^2 , \quad (9)$$
where the second term on the r.h.s. is a quantum pressure, corresponding to the kinetic energy of a Bose gas of non-uniform density. The quantity \( E_c[\rho] \) is a correlation energy, which incorporates the effects of dynamic correlations resulting from the interactions between the individual He atoms. The equilibrium configurations are obtained by minimizing the energy with respect to the density. This leads to the Hartree-type equation

\[
\left\{ -\frac{\hbar^2}{2m} \nabla^2 + U[\rho, r] \right\} \sqrt{\rho(r)} = \mu_4 \sqrt{\rho(r)},
\]

where \( U[\rho, r] \equiv \delta E_c/\delta \rho(r) \) acts as a mean field, while the chemical potential \( \mu_4 \) is introduced in order to ensure the proper normalization of the density to a fixed number of particles \( N \).

The density dependence of the correlation energy is parameterized in a phenomenological way, by choosing a functional form compatible with basic physical requirements and fixing a few parameters in accordance with the known properties of the bulk liquid. The detailed form of \( E_c \) is given in Appendix \( \text{B} \). The DF theory is particularly suitable for the calculation of the density profiles of relatively large droplets. In fact, the density functional of Ref. \( \text{[6]} \) is accurate, by construction, in the limit of the uniform liquid. It also has been tested in the opposite limit of small clusters with 20 – 70 atoms \( \text{[12]} \) for which it provides density profiles in close agreement with \( \text{ab initio} \) Monte Carlo calculations.

The density profiles shown in the upper part of Fig. \( \text{3} \) are obtained by solving Eq. (10) for \( N = 10^3 \) to \( 10^4 \). In order to estimate the contribution to the effective integral cross section \( \sigma_{\text{eff}} \) from the outer region of the theoretical profiles, the transmission coefficient \( T(b) \) of the droplets is calculated as a function of the impact parameter \( b \) for argon and krypton using Beer’s law (see the inset in Fig. \( \text{3b} \)). For this the trajectory of relative motion is integrated along a straight path \( z \), which is a good approximation in the outer region where the density is about 1% of the core density (see below):

\[
T(b) = \exp \left( -\int_{z(b)} \sigma(E_{\text{rel}}) \rho(z) \, dz \right).
\]

The atom-atom integral cross sections \( \sigma(E_{\text{rel}}) \) were again calculated using the method given in Appendix \( \text{A} \), with the relative velocities of the experiment. Here, the more realistic
Tang-Toennies potentials \[23\] are used in place of the Lennard-Jones(12,6) potentials. The calculated transmission \(T(b)\) of the droplets is illustrated in Fig. 3b. The differences between argon and krypton are less than 1\% and can therefore be neglected. The transmission for densities larger than about 10\% of the central density is almost zero, so that the cross section is largely determined by the outer region of the droplet. Since the transmission rises very steeply the effective droplet-border is rather sharp and corresponds to a radius \((R_{\text{eff}})\) where the density has fallen to approximately 1\%. The integral cross section of the droplets is then calculated with the following equation:

\[
\sigma_{\text{eff}} = 2\pi \int_0^\infty (1 - T(b)) b \, db .
\]

\(12\)

In a subsequent step the cross sections are averaged over the distribution in the number of atoms \(P(N)\). The calculated results for the effective radii \(R_{\text{eff}} = \sqrt{\sigma_{\text{eff}}/\pi}\), \(\sigma_{\text{eff}}\), the average relative densities \(\bar{\rho}/\rho_{\text{bulk}}\) and the 10–90\% thickness \(t\) are reported in Table II. The average relative densities \(\bar{\rho}/\rho_{\text{bulk}}\) in Fig. 2 (filled diamonds) lie between 0.64 and 0.8 and are somewhat larger than the experimental values by about 10–25\%. This discrepancy suggests that the theoretical surface thicknesses are somewhat smaller than the experimental values.

A simple model calculation can be used to check the effect of the surface thickness on the integral cross section. Since all theoretical calculations predict the core density of these large droplets to be equal to the bulk density, the density profile can be rather realistically described by a simple analytic function \[25,26\]:

\[
\rho(r) = \frac{\rho_{\text{bulk}}}{2} \left( 1 - \tanh \left( \frac{2r - R}{g} \right) \right) ,
\]

\(13\)

where \(R\) indicates the point where the density is reduced to 50\% of the central density and \(g\) is a parameter controlling the surface thickness. By relating the density profile to the integral cross section using the same procedure described in Section III an experimental surface thickness is estimated by fitting the experimental cross section for the different droplets. The resulting values of \(t\) are shown in Fig. 4. We obtain a mean 10–90\% thickness of 6.4 ± 1.3 Å, which is somewhat larger than the results of the DF calculation (Table II).
but still within the error limits. One has to stress here that the results are dependent on
the model used for $\rho(r)$; in particular, it assumes a symmetric density profile. Most of the
predicted density profiles are however slightly asymmetric, with a steeper slope in the outer
part of the surface. By using an asymmetric profile, with an asymmetry similar to the
one of the DF profiles in Fig. 3, the estimated surface thickness is about 1–2 Å larger than
the values shown in Fig. 4. This would increase the discrepancy between the experimental
estimates and the theoretical surface thickness.

IV. DISCUSSION

As discussed above it appears as if the experimental surface thickness is somewhat larger
than the values predicted by the DF theory. This is indicated by the 10–25% differences in
the average densities in Fig. 2 and by the larger widths obtained using model profiles similar
to Eq. (13). Since these differences are greater than the experimental uncertainties of 5%,
we consider here possible sources of error.

The possibility that the measured droplet atom numbers $N$ are too small, which would
shift the mean effective density to smaller values, seems unlikely. This would require a
momentum transfer in the deflection experiment even larger than the momentum $p_{sec}$ of
the impinging secondary atom. For elastic backscattering of the atoms in central collisions
from a hard sphere the momentum transfer would indeed be $2p_{sec}$. The momentum transfer
averaged over all impact parameters for elastic scattering from a hard sphere is, however,
equal to the momentum of the incoming particle $p_{sec}$. Thus even in the unlikely case of elastic
recoil scattering, which in view of the large mass of the argon and krypton atom compared
to that of the He atom seems very unlikely and is not even expected for the scattering from
a liquid helium surface [8], the same results would still hold. Recoil scattering would also
be in contradiction to the observation that the secondary gas is captured by the He droplets
[19, 20]. Another possibility is a backward directed vaporization of helium induced by the
impinging secondary gas atom, as proposed by Gspann [27]. According to ordinary fluid

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dynamics this would also seem implausible, since the impact speeds are greater than the velocity of first sound of about $240 \text{ m/s}$ \[27\] and therefore a conical Mach shock should be created which degenerates in a spherical sound wave. This sound wave would more probable induce vaporization in forward direction which would tend to shift our results to greater droplet atom numbers $N$ and would therefore increase the discrepancy with the theoretical results.

The comparison between measured cross sections and DF calculations is affected by at least two possible sources of problems. The first concerns the form and extent of the density profiles, and the second the overall shape of the droplets. The effect of different density profiles was already discussed in the previous section and has a noticeable effect. The 10–90% surface thickness of the density profiles from the DF calculations is $t \approx 5.7 \text{ Å}$. This value is slightly less than other estimates. For example, previous DF calculations for a planar free surface \[28\] gave $t \approx 7 \text{ Å}$, while from \textit{ab initio} calculations on small clusters \[13–18\] values between 6 and $7 \text{ Å}$ are predicted. Some experimental and theoretical results for the surface thickness of the bulk liquid and droplets are summarized in Table III.

The second possible reason why the measured cross sections are larger than the calculated ones from DF could be that the helium droplets are not spherical, as assumed in the analysis, but have the shape of an oblate ellipsoid. A flattening of the droplets does not seem unfounded, due to the large angular momentum of several $1000 \hbar$ which some of the droplets may have as a result of collisions with the residual gas before arriving in the scattering region. A simple estimate indicates that in view of the large mass of the droplet, the resulting rotation speed is slow compared with the colliding secondary beam atoms. Thus the effect of a possible deformation on the experimental results can be estimated from the calculated geometrical cross section of a rotational ellipsoid averaged over all orientations. In this estimate vibrational distortions, which at the low temperature should only have small amplitudes, are neglected.

The radius of the ellipsoid is assumed to have the length $a$ and the radius of the axis of rotational symmetry is given by $b$. For $a > b$ one gets an oblate and for $a < b$ a prolate
ellipsoid. The geometrical cross section $\sigma_{\text{ell}}$ under an angle of view $\omega$ can be written as (see Appendix C):

$$
\sigma_{\text{ell}} = \frac{4}{3} \pi a \sqrt{\frac{b^4 + a^4 \tan^2 \omega}{b^2 + a^2 \tan^2 \omega}} \sin \left( \omega + \arctan \left( \frac{b^2}{a^2 \tan \omega} \right) \right).
$$

(14)

The effective cross section is obtained by averaging numerically over all orientations,

$$
\bar{\sigma}_{\text{ell}} = \frac{1}{2} \int_0^\pi d\omega \sin(\omega) \sigma_{\text{ell}}(\omega).
$$

(15)

The resulting mean cross section is shown as a function of the ratio of the radii $b/a$ in Fig. 3, scaled to the cross section of a sphere with the same volume. For a real droplet with lower density at the surface the effect will be even greater, since the surface area of the ellipsoid is larger. To explain the difference of 5–10% between the DF results and the experiments, oblate shaped droplets with a value of $b/a \approx 0.6$ or prolate droplets with $b/a \approx 2.5$ would be required. Oblate shapes with such small average $b/a$ ratios seem unlikely in view of the relatively small fraction ($\approx 15\%$) of the incident beam droplets which undergo collisions with the residual gas. Thus we conclude that even in the very unlikely case that the droplets are somewhat distorted this has little effect on the measured surface thickness.

V. CONCLUSIONS

By using a combination of scattering techniques (deflection and attenuation), we have shown that it is possible to measure the average densities of large Helium droplets. These results have been compared with density functional calculations which have been carried out for the large droplets with $N = 10^3$ to $10^4$ Helium atoms studied in the experiment. Overall agreement can be considered to be satisfactory but on closer examination the predicted effective integral cross sections are too small be about 10–20% and the densities too large by about 10–25%, which is larger than the errors which are estimated to be about 5%. An attempt to fit the experimental data using a symmetric model density profile yields a value for the 10–90% thickness of $t = 6.4 \pm 1.3\text{Å}$ which is larger than the density functional value.
of 5.7 Å and is consistent with the differences in the densities. If an asymmetric profile is assumed 1–2 Å larger thicknesses are obtained which would increase the discrepancy. The assumptions made in the analysis were critically examined and an explanation for the small discrepancy cannot be provided. As discussed in connection with Table II the situation is similar to the surface of the bulk where experiments also yield thicknesses which are larger than predicted by most theories.

The experimental method has also been recently applied to $^3$He droplets [29] and there the average densities relative to the bulk are even smaller than found here for $^4$He, a trend which is consistent with recent Thomas-Fermi theoretical calculations [30].

In the future it is conceivable that the present experimental method can be further improved to provide a sensitive quantitative probe of the outer surface region of large droplets. Here it is interesting to speculate what effect the large condensate fraction in this outer region [11] would have on the cross section. One can also explore the existence of a Landau velocity below which the interaction should disappear, as it happens in the bulk liquid at 57 m/s. Experiments under such conditions are now possible [31] and are envisaged in the future.

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APPENDIX A: INTEGRAL CROSS SECTION

The quantum mechanical differential cross section for elastic scattering from the secondary beam gas atoms was calculated using a partial wave expansion method [24] in order to estimate the effect of the forward diffraction contribution on our results. The following
spherical model potential obtained by integrating the Lennard-Jones 12-6 potential between the scattering atom and the He atoms of the droplet was assumed \[32\],

$$V_N(r) = \frac{4N\varepsilon\kappa^6}{(r^2 - R^2)^3} \left[ \kappa^6 \left( \frac{r^6 + \frac{21}{5}r^4R^2 + 3r^2R^4 + \frac{1}{3}R^6}{(r^2 - R^2)^6} - 1 \right) \right]; \ r > R. \quad (A1)$$

The integration is simplified by assuming the He atoms to be homogeneously distributed within a sphere of radius \(R\). Here the effective radius \(R\) corresponds to the radius \(R_N = r_0N^{1/3}\) of a droplet with \(N\) atoms reduced by the effective radius \(r_0\) of one atom in the same droplet, i.e., \(R = R_N - r_0 = (N^{1/3} - 1)r_0\). Estimates of the effective radius \(r_0\) of one helium atom in a droplet can be extracted from quantum many-body calculations. For instance, Pandharipande et al. \[16\] reported the value \(r_0(N) = 2.24 + 0.38N^{-\frac{1}{3}} + 2.59N^{-\frac{2}{3}}\). The parameters \(\varepsilon\) and \(\kappa\) are such that, in the limiting case \(N = 1\), the potential \(V_N\) reduces to the two-body Lennard-Jones He-Kr or He-Ar potential:

$$V_1(r) = 4\varepsilon \left[ (\kappa/r)^{12} - (\kappa/r)^6 \right] \equiv \varepsilon \left[ \left( \frac{R_m}{r} \right)^{12} - 2 \left( \frac{R_m}{r} \right)^6 \right], \quad (A2)$$

where \(R_m = 2^{1/6}\kappa\). We use \(\varepsilon = 2.67\) meV and \(R_m = 3.70\) Å for He-Kr, and \(\varepsilon = 2.59\) meV and \(R_m = 3.40\) Å for He-Ar \[33\]. Although the inner repulsive potential is not realistic for central collisions with the strongly absorbing liquid core of the He droplet, the potential Eq. (A1) should be a good approximation for describing the glancing collisions, which are the relevant ones in determining the elastic contribution to the integral cross section.

The integral cross section of real He droplets consists of the elastic cross section \(\sigma_e\) resulting from diffraction in large impact collisions and the absorption cross section \(\sigma_a\) in more central collisions. In the calculations with the hard core potential Eq.(A1) the angularly distributed isotropic part of the differential cross section takes account of the effect of the absorption cross section \(\sigma_a\). In our case, where the de Broglie wavelength of relative motion \(\lambda\) is much smaller than the droplet radius, all particles with impact parameter smaller than approximately \(R_N\) are expected to be absorbed. The contribution of particles with angular momentum \(l\) to the cross section is given by \(\sigma_l = (2l + 1)\pi\lambda^2\) \[34\]. Then the integral cross section is given approximately by:
$$\sigma \simeq \sum_{l=0}^{\infty} (2l + 1) \pi \lambda^2 + \sum_{l=R_N/\lambda}^{\infty} \sigma_{el,l} \simeq \pi R_N^2 + \sigma_e.$$  \hspace{1cm} (A3)

The part $\sigma_e$ was calculated quantum mechanically exactly for the potential Eq. (A1). Since the calculated elastic scattering is found to be sharply peaked in the forward direction its contribution to the integral cross section can be neglected for center of mass angles $\vartheta$ larger than about $5^\circ$. The laboratory scattering angle $\Theta$ is in the case of $m_{\text{drop}} \gg m_{\text{sec}}$ related to $\vartheta$ by

$$\Theta(\vartheta) = \frac{m_{\text{sec}}}{v_{\text{drop}}(m_{\text{sec}} + m_{\text{drop}})} \left\{ v_{\text{rel}} \sin \left[ \arcsin \left( \frac{v_{\text{sec}} \sin \beta}{v_{\text{rel}}} \right) + \vartheta \right] - v_{\text{sec}} \sin \beta \right\},$$  \hspace{1cm} (A4)

where $v_{\text{rel}}$ is the relative speed of the two beams and $\beta$ is the angle between them. Since the mass $m_{\text{drop}}$ of the primary beam droplets is much larger than the mass $m_{\text{sec}}$ of the secondary gas atoms the angle $\Theta$ turns out to be very small. For example a center of mass angle $\vartheta = 5^\circ$ corresponds to laboratory angles $\Theta$ of about 1 mrad for droplets with 1000 atoms, which is comparable with the angular resolution of our apparatus.

The theoretical differential cross sections were convoluted with the angular distributions $S(\Theta)$ of the incident beam (beam profiles). These latter angular distributions were measured with the same method as described in Section [IIA] with the secondary beam blocked with a beam flag located in the scattering chamber. In this arrangement the residual gas pressure and its effect on the beam profile is the same as in the scattering experiment. The predicted effective integral cross section is then given by:

$$\sigma_{\text{app}}^{\text{int}} = 2\pi \int_{\vartheta_{\text{app}}}^{\pi} d\vartheta \sin(\vartheta) \left\{ \int_{-\pi}^{\pi} d\zeta \left( \frac{d\sigma(\vartheta - \zeta)}{d\omega} \right)_{\text{th}} s(\zeta) \right\},$$  \hspace{1cm} (A5)

where the $\vartheta_{\text{app}}$ represents the effective geometrical center of mass resolution given by the slit in front of the scattering center and in front of the detector and $s(\vartheta)$ is the measured angular profile of the unscattered beam transformed into the center of mass system. The quantity $(d\sigma/d\omega)_{\text{th}}$ is the calculated elastic differential cross section. Fortunately the resulting value of $\sigma_{\text{app}}^{\text{int}}$ turns out to be very close to the classical cross section $\sigma_{\text{class}} = \pi R_N^2$; the two cross sections agree to within 1% for droplets with 1000 atoms and for larger droplets the
agreement is even better. Thus the corrections discussed above justify the assumption that
the classical cross section is measured.

APPENDIX B: DENSITY FUNCTIONAL

The explicit expression of the correlation energy $E_c$, entering the density functional (9),
is given by [6,12]:

$$
E_c[\rho] = \int dr \left\{ \frac{1}{2} \int dr' \rho(r)V_l(|r-r'|)\rho(r') + \frac{c_2}{2}\rho(r)(\overline{\rho_h}(r))^2 + \frac{c_3}{3}\rho(r)(\rho_h(r))^3
\right.
- \frac{\hbar^2}{4m\alpha_s}\int dr' F(|r-r'|) \left( 1 - \frac{\rho(r)}{\rho_{0s}} \right) \nabla \rho(r) \cdot \nabla \rho(r') \left( 1 - \frac{\rho(r)}{\rho_{0s}} \right) \right\} .
$$

(B1)

The two-body interaction $V_l$ is the Lennard-Jones interatomic potential, with the standard
parameters $\alpha = 2.556 \ \text{Å}$ and $\varepsilon = 10.22 \ \text{K}$, screened at short distance ($V_l \equiv 0$ for $r < h$,
with $h = 2.1903\text{Å}$). The two terms with the parameters $c_2 = -2.411857 \times 10^4 \ \text{K} \ \text{Å}^6$
and $c_3 = 1.858496 \times 10^6 \ \text{K} \ \text{Å}^9$ account phenomenologically for short range correlations between
atoms. The weighted density $\overline{\rho_h}(r)$ is the average of $\rho$ over a sphere of radius $h$ centered in
$r$. The last term, depending on the gradient of the density in different points, is introduced
to reproduce the static response function in the roton region. The function $F$ is a simple
Gaussian, $F(r) = \pi^{-3/2}\ell^{-3}\exp(-r^2/\ell^2)$ with $\ell = 1 \ \text{Å}$, while $\alpha_s = 54.31 \ \text{Å}^3$ and $\rho_{0s} = 0.04$
$\text{Å}^{-3}$.

APPENDIX C: MEAN GEOMETRICAL CROSS SECTION OF A ROTATIONAL

ELLIPSOID

A rotational ellipsoid is assumed with the axis of rotational symmetry $b$ parallel to the
z-axis and the other two radii $a$: 

$$
\frac{x^2}{a^2} + \frac{y^2}{b^2} + \frac{z^2}{b^2} = 1 .
$$

(C1)

This can be written in spherical coordinates:
\[ r(\Theta, \Phi) = \frac{b}{1 - \epsilon^2 \sin^2(\Theta)}; \quad \epsilon = \frac{\sqrt{a^2 - b^2}}{a} < 1. \] 

(C2)

Since the visible cross section \( \sigma_{\text{ell}} \) of the ellipsoid from a viewing point with a polar-angle \( \omega \) 
does not depend on the azimuth-angle \( \Phi \), the problem can be reduced to the x-z-plane. The 
visible cross section \( \sigma_{\text{ell}} \) is given by the projection of the cutting plane through the ellipsoid 
which is defined by the tangents with angle \( \omega \). Since every cutting plane of an ellipsoid is 
an ellipse, the area of the cutting plane \( \sigma_s \) can written as:

\[ \sigma_s(\omega) = \pi ar_s(\omega). \] 

(C3)

The projection on a plane perpendicular to the viewing direction is given by:

\[ \sigma_{\text{ell}}(\omega) = \sigma_s(\omega) \cos(\omega - \Theta_s). \] 

(C4)

To find the vector \( r_s = \left( \begin{array}{c} x_s \\ \pi - \Theta_s \end{array} \right) = \left( \begin{array}{c} x_s \\ z_s \end{array} \right) \) lying in the cutting plane the equation of a tangent 
at the ellipse has to be calculated

\[ 1 = \frac{xx_s}{a^2} + \frac{zz_s}{b^2} \] 

(C5)

which yields

\[ z = \frac{b^2}{z_s} - \frac{b^2 x_s}{a^2 z_s} x. \] 

(C6)

The tangent must be parallel to the viewing direction \( \omega \)

\[ \cot(\omega) = \frac{dz}{dx} = -\frac{b^2 x_s}{a^2 z_s}. \] 

(C7)

With equation (C1) follows

\[ x_s^2 = \frac{a^4 \cot^2(\omega)}{b^2 + a^2 \cot^2(\omega)}. \] 

(C8)

and for \( z_s \) and \( r_s \)

\[ z_s^2 = \frac{b^4}{b^2 + a^2 \cot^2(\omega)}, \] 

(C9)

18
\[ r_s^2 = x_s^2 + z_s^2 = \frac{b^4 + a^4 \cot^2(\omega)}{b^2 + a^2 \cot^2(\omega)}. \]  

(C10)

From Eqs. (C8) and (C9) \( \Theta_s \) can be calculated:

\[ \tan(\Theta_s) = \frac{z_s}{x_s} = \frac{b^2}{a^2 \cot(\omega)}. \]  

(C11)

The visible cross section of an ellipsoid follows from Eqs. (C3) and (C4)

\[ \sigma_{\text{ell}}(\omega) = \pi a \sqrt{\frac{b^4 + a^4 \cot^2(\omega)}{b^2 + a^2 \cot^2(\omega)}} \cos \left[ \omega - \arctan \left( \frac{b^2}{a^2 \cot(\omega)} \right) \right]. \]  

(C12)

The mean cross section is derived by integration over \( \omega \)

\[ \overline{\sigma_{\text{ell}}(\omega)} = \frac{1}{2} \int_0^\pi d\omega \sin(\omega) \sigma_{\text{ell}}(\omega). \]  

(C13)
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TABLE I. Experimental results as function of source temperature ($T_0$), source pressure ($P_0$) and secondary gas. The mean number of atoms $\overline{N}$ and the half-width $\Delta N_{1/2}$ are the results of fitting the measured mass distributions from the deflection experiment with a log-normal distribution (parameters $\delta$ and $\mu$, see Eq. (1)). The mean classical integral cross section $\sigma$ is obtained by attenuation of the droplet beam with the secondary beam. From this data the mean density of the droplets $\rho$ as a fraction of the known bulk density ($\rho_{\text{bulk}} = 0.0218 \text{ Å}^{-3}$) is obtained directly.

| $T_0$ [K] | $P_0$ [bar] | sec. gas | $\overline{N}$ | $\Delta N_{1/2}$ | $\delta$ | $\mu$ | $\sigma$ [Å$^2$] | $\rho/\rho_{\text{bulk}}$ |
|-----------|-------------|----------|----------------|---------------|--------|------|----------------|-----------------|
| 24.0      | 40          | Kr       | 703            | 667           | 0.426  | 6.46 | 2266          | 0.40            |
| 22.0      | 40          | Kr       | 1700           | 1632          | 0.407  | 7.36 | 3138          | 0.59            |
| 20.0      | 40          | Kr       | 2617           | 2373          | 0.528  | 7.73 | 4519          | 0.53            |
| 18.0      | 40          | Kr       | 4700           | 4158          | 0.573  | 8.29 | 6259          | 0.58            |
| 17.0      | 40          | Kr       | 6130           | 5331          | 0.603  | 8.54 | 7108          | 0.62            |
| 16.0      | 40          | Kr       | 7741           | 6484          | 0.662  | 8.74 | 7661          | 0.70            |
| 15.0      | 40          | Kr       | 8900           | 7719          | 0.607  | 8.91 | 8540          | 0.69            |
| 13.5      | 40          | Kr       | 13000          | 11240         | 0.612  | 9.29 | 9538          | 0.85            |
| 26.0 a    | 80          | Kr       | 1460           | 1298          | 0.565  | 7.13 | 3106          | 0.51            |
| 24.0 a    | 80          | Kr       | 2700           | 2524          | 0.468  | 7.79 | 4111          | 0.62            |
| 18.0 a    | 80          | Kr       | 5260           | 4374          | 0.673  | 8.34 | 6270          | 0.65            |
| 26.0      | 80          | Ar       | 2114           | 1967          | 0.478  | 7.54 | 3431          | 0.64            |
| 24.0      | 80          | Ar       | 3103           | 2835          | 0.514  | 7.91 | 4443          | 0.64            |
| 20.0      | 80          | Ar       | 6458           | 5916          | 0.509  | 8.64 | 6615          | 0.70            |
| 18.0      | 80          | Ar       | 9487           | 8530          | 0.544  | 9.01 | 9025          | 0.67            |

a The smaller cluster number sizes found with Kr instead of Ar as scattering gas at $P_0 = 80$ bar is a consequence of a smaller nozzle with an estimated diameter of about 4µm.
TABLE II. The effective radii $R_{\text{eff}} = \sqrt{\frac{\sigma_{\text{eff}}}{\pi}}$ and cross sections $\sigma_{\text{eff}}$ obtained using Eq. (12) and DF calculations of density profiles. The effective density $\bar{\rho}$ as a fraction of the known bulk density ($\rho_{\text{bulk}} = 0.0218 \text{ Å}^{-3}$) and the 10–90% surface thickness $t$ are listed.

| $N$  | $R_{\text{eff}}$ [Å] | $\sigma_{\text{eff}}$ [Å$^2$] | $\bar{\rho}/\rho_{\text{bulk}}$ | $t$ [Å] |
|------|----------------------|-------------------------------|---------------------------------|--------|
| 1000 | 25.75                | 2083                          | 0.64                            | 5.6    |
| 2000 | 31.59                | 3135                          | 0.69                            | 5.6    |
| 3000 | 35.69                | 4000                          | 0.72                            | 5.7    |
| 4000 | 38.94                | 4763                          | 0.74                            | 5.7    |
| 5000 | 41.69                | 5459                          | 0.76                            | 5.7    |
| 6000 | 44.09                | 6106                          | 0.77                            | 5.7    |
| 7000 | 46.22                | 6713                          | 0.78                            | 5.7    |
| 8000 | 48.18                | 7292                          | 0.78                            | 5.7    |
| 9000 | 49.97                | 7843                          | 0.79                            | 5.7    |
| 10000| 51.63                | 8373                          | 0.80                            | 5.7    |
TABLE III. The 10–90% surface thickness of $^4$He$_N$ droplets and bulk liquid helium from different published experimental and theoretical works, where $N$ is the number of atoms in the droplets and $T$ the temperature (the values for $N = \infty$ refer to the planar free surface).

| Method                  | $N$     | $T$ [K] | $t$ [Å] |
|-------------------------|---------|---------|---------|
| Osborne (1989)          | $\infty$ | ellipsometry | 1.8 | 9.4 |
| Lurio et al. (1992)     | $\infty$ | X-Ray  | 1.13 | 9.2 |
|                         |         | extrapol. | 0   | 7.6 |
| Pandharipande et al. (1983) | $< 728$ | GFMC,VMC | 0 | 5.5–7.2 |
| Stringari, Treiner (1987) | $< 728$ | DF      | 0   | 8.8–9.2 |
|                         | $\infty$ | DF      | 0   | 7    |
| Sindzingre, Klein, Ceperley (1989) | 64–128 | PIMC    | 0.5–2 | $\simeq 6$ |
| Guirao, Centelles, Barranco et al. (1992) | $\infty$ | DF  | 0–4 | 6.5 (at 0.4 K) |
| Chin, Krotscheck (1995) | 20–112 | DMC    | 0   | $\simeq 6$ |
|                         | 20–1000 | HNC    | 0   | $\simeq 6$ |
| Barnett, Whaley (1995)  | $< 112$ | DMC    | 0   | $\simeq 6$ |
FIGURES

FIG. 1. Three typical measured angular distributions for a source pressure of $P_0 = 80 \text{ bar}$ and source temperatures of $T_0 = 25 \text{ K}$ (a), 20 K (b) and 17 K (c). Krypton was used as secondary beam gas. The signals with (filled circles) and without (open circles) a flag in front of the secondary beam are shown on a logarithmic scale. The weighted differences of the two signals (diamonds) with the standard deviations are shown on a linear scale. The mean number of atoms in the droplets are: a) $\bar{N} = 2602$, b) $\bar{N} = 6174$, c) $\bar{N} = 9834$. The integral cross section is determined from the attenuation of the forward peak, i.e. 0 mrad.

FIG. 2. The classical integral cross sections averaged over the measured number size distributions as functions of mean number of atoms $\bar{N}$. The empty symbols show the experimental results. The solid line with filled diamonds is the result of the DF calculations. For comparison, the mean classical cross sections of spherical droplets with constant density are indicated as dashed lines for different values of the density ($\rho_{\text{bulk}} = 0.0218 \text{ Å}^{-3}$). The different empty symbols indicate the different experimental parameters: triangle: $P_0 = 40 \text{ bar}$, Sec.-gas=Kr; square: $P_0 = 80 \text{ bar}$, Sec.-gas=Kr; circle: $P_0 = 80 \text{ bar}$, Sec.-gas = Ar.

FIG. 3. The density distributions a) calculated with a density functional method for droplets with between $N = 10^3$ and $N = 10^4$ atoms with steps of $10^3$. The corresponding transmission for a beam of krypton or argon atoms is shown in b). The effective radius $R_{\text{eff}}$ for a droplet with $10^4$ atoms is also indicated.

FIG. 4. The experimental 10–90% surface thickness of He droplets as function of the mean number of atoms $\bar{N}$ assuming the symmetric density profile (Eq. (13)) shown in the inset. The mean value of $t$ is $6.4 \pm 1.3\text{Å}$. The DF values are shown as a dotted line. The different empty symbols indicate the different experimental parameters: triangle: $P_0 = 40 \text{ bar}$, Sec.-gas=Kr; square: $P_0 = 80 \text{ bar}$, Sec.-gas=Kr; circle: $P_0 = 80 \text{ bar}$, Sec.-gas = Ar.
FIG. 5. The effective integral cross section of an ellipsoid averaged over all possible orientations in units of the cross section of a sphere with the same volume. The radius $b$ is the axis of symmetry and $a$ indicates the other radius.
Graphs showing signal and difference signal data with various temperature levels. Notations and values are as follows:

- Signal [1/s] graphs with logarithmic scales.
- Difference Signal [10^3 1/s] shown for different temperature levels:
  - $T_0 = 25$ K
  - $T_0 = 20$ K
  - $T_0 = 17$ K
- Angle [mrad] values along the x-axis.

Legend:
- Circles represent data points.
- Error bars indicate variation or uncertainty in measurements.
\[ I = \frac{I}{I_0} = \exp(-\rho(z) \sigma dz) \]
Mean Number of Atoms, $N_{[10^3]}$

Surface Thickness, $t [\text{Å}]$

$P_0 = 80 \text{ bar}, \text{Sec.=} \text{Ar}$

$P_0 = 40 \text{ bar}, \text{Sec.=} \text{Kr}$

$P_0 = 80 \text{ bar}, \text{Sec.=} \text{Kr}$

DFC
