Differences in the Detachment of Electron Bubbles from Superfluid ^{4}He Droplets versus Nonsuperfluid ^{3}He Droplets

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(Received 8 June 1998)

The rate of detachment of electron bubbles inside size-selected ^{4}He and ^{3}He droplets (10^5-10^7 atoms) has been measured. At zero electric field the lifetimes of ^{4}He droplets increase with size from 10^{-3} to 0.2 s, orders of magnitude smaller than previously predicted. With increasing fields the detachment gradually increases for ^{4}He droplets, whereas a sharp threshold was found at 1.45 kV/cm for ^{3}He droplets, independent of size. A simple *dynamical* model which accounts for superfluidity in the ^{4}He droplets versus a viscous dynamics in ^{3}He can explain the observations. [S0031-9007(98)07487-0]

PACS numbers: 67.40.Yv, 36.40.–c

It is now well established that an excess electron in bulk liquid He undergoes a type of Anderson localization transition from a conduction band to a nonconducting state in which it is surrounded by a bubble with a radius of $r_b \sim 17$ Å [1]. Jiang *et al.* [2] showed experimentally that electrons are also preferentially localized in the interior of large ^{4}He droplets with $N > 10^5$ atoms ($R > 100$ Å). These "bubblons" are unique quantum dots which open up new opportunities to explore the superfluidity of ^{4}He on a microscopic level [3]. Since the electron ground state lies about 0.1 eV above the vacuum level, the bubblons are essentially metastable; however, the mechanisms by which the bubbles escape from the droplets and from the bulk are still not completely understood. The bubble tunneling rates measured in the bulk [4] have been recently used by Northby *et al.* to predict lifetimes between 10^5 s ($N \sim 10^5$) and 10^{15} s ($N \sim 2.5 \times 10^6$) in the ^{4}He droplets [5], which are drastically greater than our measured lifetimes of $\tau \leq 0.2$ s. This discrepancy indicates a new physical phenomenon occurring in the droplets, which apparently was not taken into account in the previous calculations.

In order to resolve these issues a series of droplet beam experiments have been carried out using size-selected ^{3}He and ^{4}He droplets in the range $N \approx 10^5-10^7$ with and without an electric field. Large differences are observed in both the lifetimes without field and in the behavior with an applied field for the two isotopes, and this led us to propose a new dynamical model involving bubble oscillations within the droplet. It requires assuming a nearly unhindered motion of the electron bubble in a ^{4}He droplet. This is consistent with recent high resolution spectroscopy studies of single embedded dopant molecules, which not only provide direct evidence for temperatures of 0.37 K [6], which are sufficiently low to support superfluidity [7], but also confirm the occurrence of superfluidity [8] and a greatly reduced drag on molecular rotations in ^{4}He droplets [3]. In Table I the known bubble properties in both isotopes are summarized.

The experimental apparatus is shown schematically in Fig. 1 [9]. Helium droplets with a relative narrow velocity spread ($\Delta v/v \leq 4\%$) are produced in a high pressure (20 bar) expansion at source temperatures between 5 and 9.5 K through a 5 μm diameter 20 μm long nozzle. A highly monochromatic electron beam ($\Delta E/E \cong 0.5\%$) from a high current, $I = 0.1$ mA, homemade electron source operating at a previously established optimum electron energy of about $E_e = 22$ eV [10] was used to produce the electron bubbles. An on-line electrostatic field energy analyzer described in [9] serves as a droplet size selector with a size resolution of $\Delta N/N \approx 5\%$ and blocks out all remaining neutral droplets. An electrostatic in-line acceleration-deceleration device provides for a high electric field. A vacuum of better than 10^{-9} mbar was maintained along the droplet beam path so that less than 6% of the droplets are contaminated by a single impurity molecule [10].

In order to determine the relative number of droplets neutralized by electron detachment in the electric field, the remaining charged droplets were deflected out of the beam by the deflection field and the neutralized droplet signal S^n was measured by the pressure increase in a flux sensitive pitot tube. This was normalized to

TABLE I. Comparison of electron bubble parameters for bulk liquid ^{4}He and ^{3}He.

Bubble parameter	^{4}He II	^{3}He
Radius, r_b (Å)	17 [a,b]	20–21 [c]
Effective mass, m_b	$243 \times m_{^4\text{He}}$ [b]	$290 \times m_{^3\text{He}}$ [c]
Conduction band energy, V_0 (eV)	0.95–1.3 [a,d]	0.65–1.1 [d]
Ground state energy, V_g (eV)	0.08 [a]	0.06 [e]
He viscosity, η (μpoise)	~0 [f]	195 [g]

[a]Ref. [19]. [b]Ref. [23]. [c]Ref. [24]. [d]Ref. [25]. [e]This value was calculated assuming the square-well-potential model with the bubble radius of 20.5 Å. [f]The viscosity of superfluid ^{4}He at 0.37 K is generally regarded as being zero since the normal fraction is less than 2.1×10^{-6} [26]. [g]Ref. [12]

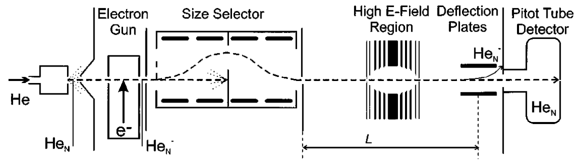

FIG. 1. Schematic diagram of the apparatus. The actual apparatus is partitioned into seven differentially pumped vacuum chambers (not shown), and the beam path is surrounded with liquid N_2 shields (not shown) to reach a residual gas pressure better than 10^{-9} mbar. The total beam path from the nozzle to pitot tube is ~200 cm.

the signal without deflection S_0^- which corresponds to the total number of negatively charged droplets exiting from the size selector to give the detachment ratio $n = S^n/S_0^-$. Without field the spontaneous detachment ratio n_0 was measured and the droplet lifetime τ was estimated from the exponential rise of n_0 with the detachment time t: $n_0 = 1 - \exp(-t/\tau)$. In the field-free measurements, $t = L/v$ was varied by mechanically changing the distance L from 23 to 67 cm (see Fig. 1). The lifetimes for a given size N were independent of the source temperature, independent of the electron energy [10], and independent of the electric field used to size select the droplets.

Figure 2 shows examples of the measured detachment ratios n as a function of the applied field E for various droplet sizes N for both ^{4}He and ^{3}He droplets. Whereas the field-induced electron detachment ratio increases rather smoothly with size N in the case of ^{4}He, the detachment ratios for ^{3}He droplets exhibit a sharp threshold at $E_{\text{th}} = 1.45 \pm 0.05$ kV/cm. This behavior was confirmed for 15 different sizes between 10^6 and 2×10^7 atoms. Without applied field the measured ratio n_0 is significantly above zero for ^{4}He droplets and the lifetimes shown in the inset in Fig. 2 were determined from measurements with various decay lengths L. For ^{3}He droplets n_0 was always smaller than the present experimental sensitivity, leading to a lower lifetime limit of $\tau > 2$ s.

To explain these results a simple one-dimensional dynamical model [11] is proposed in which the helium droplet with radius R is approximated by a slab of helium between $-R$ and $+R$. The bubble dynamics at a distance x from the droplet center can be described by the Langevin equation:

$$m_b \ddot{x} + \frac{dV(x)}{dx} = f_D(x,t) + f_T(x,t), \qquad (1)$$

where f_D is the drag force and f_T is the fluctuation force exerted by the bubble environment. Since the bubble motion was found to be largely insensitive to the actual shape of the bubble potential, the image charge potential was approximated with a harmonic potential. The frequency of the undamped oscillator was calculated to be $\omega_0 \sim 10^8$–10^9 s^{-1}.

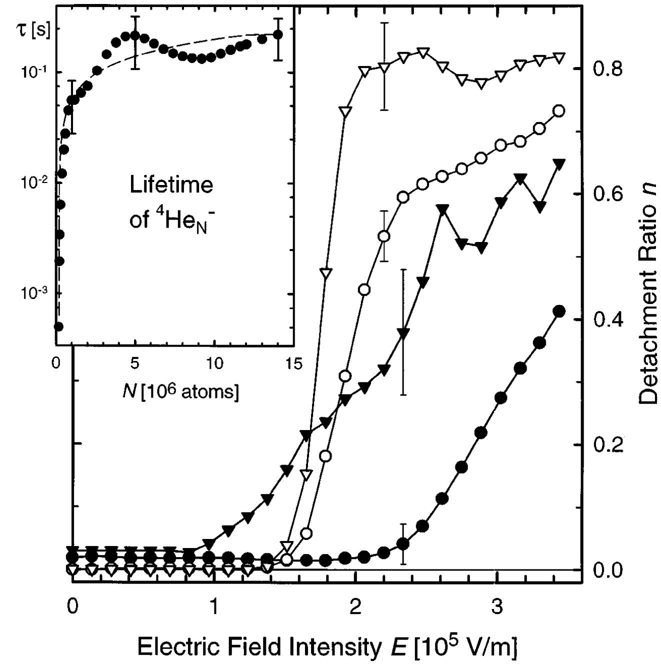

FIG. 2. The detachment ratio n as a function of the electric field intensity E for ^{3}He droplets (open symbols) and ^{4}He droplets (full symbols) for various droplet sizes $N \approx 5 \times 10^6$ (triangles) and 14×10^6 (circles). The inset shows the measured droplet lifetimes τ for ^{4}He droplets without electric field as a function of droplet size N. The dashed line corresponds to the fit with the present model, Eq. (4).

Since ^{3}He is a normal fluid with very high viscosity $\eta = 195$ μpoise [12] at the droplet temperature of 0.15 K [13], the drag force on the bubble can be described by Stokes' law [14] $f_D = -4\pi\eta r_b \dot{x}$. Thus, the characteristic damping time $\tau_D = m_b/4\pi\eta r_b \approx 6 \times 10^{-12}$ s is very short compared to the oscillation period $\tau_0 = 2\pi/\omega_0 \approx 10^{-8}$–$10^{-7}$ s, and the bubble undergoes an overdamped motion towards the central equilibrium position. The fluctuation term f_T leads to the Boltzmann probability distribution of the thermalized bubble about the droplet center. Thus, the static theory of Northby *et al.* [5] can be used to calculate the ^{3}He droplet lifetimes, which are much longer than the experimental limit of 2 s.

On the other hand, in superfluid ^{4}He, since the maximum bubble velocity $v_b \approx 20$ ms^{-1} is much less than the Landau velocity $v_L = 58$ ms^{-1}, the drag on the negative ion is negligibly small as demonstrated previously in the bulk at higher pressures [15,16]. The small remaining scattering of thermal excitations [14] can be largely neglected in the superfluid ^{4}He droplets since the volume excitations are suppressed in the inside by finite size effects [17,18]. With the damping and the driving term zero, Eq. (1) is the equation of a free harmonic oscillator. The bubble thus bounces back and forth between the surface barriers of a width w through which the electron eventually tunnels out with the tunneling probability $q_t(w) = e^{-\alpha w}$, where $\alpha = \sqrt{8m_e\Delta E}/\hbar$ (ΔE is the barrier height ≅1.05 eV) [4]. In this case the bubble lifetime can be roughly estimated as

$$\tau = \frac{\pi}{\omega_0 q_t(w)}, \qquad (2)$$

where $w = d - r_b$, and d is the distance from the surface, where the bubble was initially produced. The measured lifetimes were fitted with this expression using d as the fitting parameter (dashed line in Fig. 2), leading to $d = 33 \pm 2$ Å. This distance is roughly consistent with the expected range of 22 eV electrons in liquid ^{4}He. It also agrees with the position of the outer barrier maximum $\phi_B = 3.27$ meV in bulk ^{4}He obtained from a density functional (DF) calculation by Ancilotto and Toigo [19]. The bubbles produced with $d < 23$ Å are expected to burst immediately [19] and those with $d > 33$ Å are less probable and would tend to have much greater lifetimes. Recently, Jortner and Rosenblit have independently proposed a very similar model for the bubble dynamics in superfluid ^{4}He droplets [20].

In view of the different types of bubble motion in the two isotopes, large differences are also expected in an electric field. As illustrated in Fig. 3(b), the bubble in ^{3}He will move to one side to a position determined by the outer barrier ϕ_B. The equilibrium position of the bubble in fields up to 3 kV/cm is estimated to be considerably greater than 33 Å from the surface and the tunneling rate is still negligible. With increasing field, however, the bubble can be extracted directly without tunneling when the slope of the potential near the droplet edge approaches zero. The maximum slope of the potential curve inside a ^{4}He droplet can be estimated from DF calculations [19] to be about 10 kV/cm. For ^{3}He droplets a substantially lower barrier closer to the experimental value of 1.45 kV/cm is expected in view of (i) the lower dielectric constant, (ii) the larger bubble radius in ^{3}He, and (iii) the greater width of the 10%–90% density falloff at the surface which is ~9 Å instead of ~7 Å in ^{4}He [21]. In the ^{4}He droplets the oscillatory motion is expected to persist in the presence of the field. As shown in Fig. 3(d), both the central position and the distance at which the tunneling occurs are shifted to the edge of the droplet. The lifetime will be shortened due to the field-induced reduction in d, which leads to the increase of the tunneling rates with the field.

Additional experimental support for the model is provided by experiments in which the droplets were doped with single O_2 molecules. As an efficient scavenger the electrons are trapped by the molecule [1] while at the same time the ground state is shifted to negative energies due to the electron affinity of O_2 of about 0.44 eV. In ^{4}He droplets the n_0 ratio dropped to zero, consistent with the resulting inhibition of tunneling. Also, in the electric field the detachment ratio in ^{4}He droplets also decreased. In ^{3}He droplets no significant effect on the threshold behavior was observed, which is consistent with the assumption that tunneling does not contribute to detachment from ^{3}He droplets.

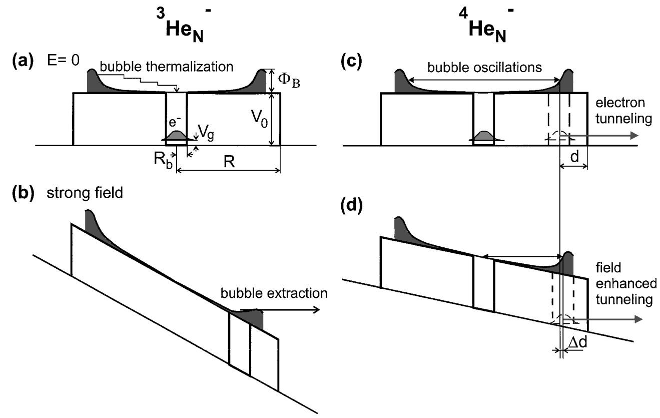

FIG. 3. Qualitative picture of the electron detachment from ^{3}He and ^{4}He droplets without an electric field (a) and (c), respectively, and with an electric field (b) and (d), respectively.

Thus, despite its simplicity, this one-dimensional dynamical model is able to explain qualitatively all the experimental observations and provides new evidence for frictionless bubble motion consistent with superfluidity in finite size ^{4}He droplets. A more quantitative comparison with the measured detachment ratios in the field would require three-dimensional density functional calculations of the bubble potential, which have not yet been extended to droplets and to the ^{3}He isotope. Also, a more precise estimate should include the effects of droplet surface excitations, and Bernoulli forces on the bubble motion [22].

We thank Joshua Jortner, Jan Northby, and Michael Rosenblit for discussions on the theory, and John Close for commenting on the manuscript. We also profited from discussions with Russ J. Donnelly, Jan Harms, Wilfried Schoepe, Domenico Stranges, Vladislav Surko, Flavio Toigo, and William Zimmermann. M. F. gratefully acknowledges the support of the Alexander von Humboldt Foundation and the Max-Planck Society.

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