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Evidence for Condensation of He^3 Atoms on the Surface of Bubbles in Liquid He^4†

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A phenomenological model is presented to explain the reduction in the critical velocity for vortex-ring creation by negative ions in liquid helium when small concentrations of He^3 impurities are added. The model is based on the assumption that the surface state for a He^3 atom has a lower energy than the bulk state. This results in an enhanced concentration of He^3 atoms on the surface of the negative ions. This change in the surface affects the flow of liquid helium in the boundary layer around the ion. The effects of the difference in the surface of the positive and negative ions on the critical velocity is discussed in both the continuous and instantaneous vortex-ring creation models.

The creation of vortex rings in liquid helium was first discovered by Rayfield and Reif.[1] The critical velocity of the ion for vortex-ring creation has since been studied by Rayfield.[2,3] Recently Rayfield[4] reported a reduced critical velocity for vortex-ring formation by negative ions in the presence of He^3 impurities. The corresponding critical velocity for positive ions was found to be independent of He^3 concentration. The two species of ions are considered to differ drastically in structure. The commonly accepted models for the ions are as follows: The positive ion consists of a hard core of solid helium[5]; and the negative ion is an electron surrounded by a bubble.[6] The experimentally determined radii of the ions are 6.4 Å for the positive ion[7] and 15–21 Å for the negative ion[7–9] at zero external pressure. Rayfield reports the following results. The critical velocity is inversely proportional to the pressure-dependent radius of the bubble. The critical velocity for the negative species is 34 m/sec at zero pressure while the corresponding velocity for the positive ion is 40 m/sec and is independent of pressure. The negative-ion critical velocity is temperature- and concentration-dependent in small concentrations of He^3. At an atomic concentration of He^3 greater than 10^{-4} the critical velocity is independent of concentration at 0.3 °K. Its magnitude is 31 m/sec at zero pressure. The pressure dependence of the critical velocity is reduced as the concentration is increased and the temperature is lowered. We believe Rayfield concludes correctly that these results indicate that the structure of the negative ion is influenced by the presence of He^3 impurities. The structure can be changed by the condensation of He^3 atoms onto the surface of the bubble. This will have three effects. It will change the surface tension and therefore the radius of the ion. It will affect the boundary conditions between the ion and the superfluid. There will be an effective increase in the ionic radius (one atomic dimension for a completely filled shell of He^3).

The zero-point energy of a He^3 atom in liquid He^4 is large because of its small mass. The zero-point energy of a He^3 atom in liquid He^4 at zero external pressure is 4.32 °K greater than the corresponding energy of a He^4 atom. We assume that the zero-point energy of the surface states is less if the He^3 atom can take advantage of a slightly increased volume at the surface. A resultant decrease in density at the surface also has the effect of reducing the repulsive interaction of the electron with the liquid. If the entire surface of approximately 200 atoms is replaced by He^3 atoms, the gain in energy per atom would be

$$\Delta E = (4\pi/N_S)\Delta V_0 \int_R^{R+d} \psi^*(r)\psi(r)r^2 dr \simeq 250 \text{ m°K}. \qquad (1)$$

Here R is the bubble radius taken to be 15 Å, d is the interatomic spacing, ΔV_0 is the difference between the barrier height for an electron at liquid He^4 and He^3 densities, N_S is the numbers of surface atoms, and $\psi(r)$ is the wave function of the bubble electron. From these energy gains must be subtracted the loss in van der Waals's energy at the surface.

Andreev[10] has calculated the density of He^3 atoms on the surface, n_S, for a surface state of energy ϵ below the bulk state for He^3. For dilute solutions he derives

$$n_S = C(\hbar\rho/m)(\mu/M)(2\pi/MT)^{\frac{1}{2}} e^{\epsilon/T}, \qquad (2)$$

$$\text{for} \quad C \ll (m/\rho)(mT/\pi\hbar^2)^{\frac{3}{2}} e^{-\epsilon/T}. \qquad (3)$$

Here C is the bulk concentration of He^3 atoms, ρ is the density of the liquid, m is the mass of the He^4 atom, and μ and M are, respectively, the effective mass of the He^3 atom on the surface and in the bulk. He derives the following formula for the decrease in surface tension:

$$\Delta\alpha = -C(\hbar\rho/m)(\mu/M)(2\pi T/M)^{\frac{1}{2}} e^{\epsilon/T}. \qquad (4)$$

From surface-tension data in He^3-He^4 solutions[11] he concludes that

$$\epsilon \simeq 3.1 °K, \quad \mu \simeq M.$$

It should be noted, however, that Esel'son *et al.*[12] explain the same results without the aid of Eq. (4). We expect ϵ to be pressure-dependent and also dependent on n_S for large n_S when He^3-He^3 interactions occur on the surface.

The radius of the bubble is inversely proportional to the $\frac{1}{4}$ power of the surface tension. The decrease in critical velocity at zero pressure might be explained by a decrease in the surface tension and a resultant increase in the radius of the bubble. The increase in surface tension given by Eq. (4) is, however, negligible at concentrations limited by Eq. (3). The surface-tension pressure is a few atmospheres, and any change in surface tension should have a negligible effect at higher pressures.

A change in the surface structure, however, might have a large effect on the critical velocity. We agree with Rayfield's suggestion[2] that the difference in the product of the critical velocity V_c and the ionic radius R for the two species is due to the boundary conditions at the surface of the ions ($V_c^-R^- \geq 2V_c^+R^+$). We might suggest that the condensation of He^3 atoms onto the surface of the bubble will change the boundary conditions in a continuous manner from those of a bubble to those of a solid surface as n_S varies from zero to unity. A small density of He^3 surface atoms might be effective in partially restricting the motion of all of the atoms in the surface layer. The maximum reduction in the critical velocity measured by Rayfield at zero pressure was ~10%. The larger decrease at higher pressures might be explained by a pressure-dependent ϵ.

We do not understand very well how the boundary conditions affect the critical velocity. The flow of the superfluid is potential flow and independent of the nature of a smooth surface.[13] In a normal fluid, however, a finite value of viscosity requires that the tangential velocity component of the liquid be continuous across the surface of the ion, and the nature of the surface is important in determining the flow pattern of the fluid. In laminar flow the boundary conditions require that the velocity of the fluid at the surface of a solid sphere be equal to the velocity of the sphere.[14] The corresponding boundary conditions at the surface of a bubble require the following[15]: (1) The radial velocity of the fluid relative to the drop must vanish, and (2) the (r, θ) component of the viscosity stress tensor must vanish. The resultant velocity of the fluid in the boundary layer 90° from the direction of motion is equal to *one-half* of the bubble velocity.

Although the dilute normal fluid cannot be adequately described by viscous hydrodynamics, there is evidence that phonon scattering may depend on the nature of the ion. The ion-phonon scattering cross section for the negative ion is larger by a factor of 25 than the corresponding cross section of the positive ion.[16,17] If the normal fluid plays a role in the creation of vortex rings, then the nature of the surface of the ion may be important in determining the critical velocity for vortex-ring creation.

It is possible that the normal fluid plays no role in the creation of a vortex ring. The ion can create a vortex ring in a time comparable to h/E when it attains a velocity[18]

$$V_c = E/P + P/2m^* . \tag{5}$$

Here E and P are the energy and momentum of the vortex ring and m^* in the effective mass of the ion. Reif[19] shows that this critical velocity should qualitatively be inversely proportional to the ionic radius as observed for the negative ion. We will comment on a means by which the boundary conditions might affect the critical velocity in this process. The energy and momentum of a free vortex ring with a radius approximately equal to the ionic radius are greater than the kinetic energy and momentum of the ion at its critical velocity. However, these quantities should be reduced if the vortex ring, or a segment of it with ends attached to the ion, is created very near to the surface of the ion. Here the ion displaces much of the high-velocity fluid near the core of the ring. If the ring is created near the negative ion, there is an additional energy term resulting from the deformation of the surface by the Bernoulli pressure. We estimate here the deformation energy of a free helium surface due to a vortex line located a distance a beneath the surface.

The coordinate system used is shown in Fig. 1. The surface lies in the $y = 0$ plane with a vortex line of circulation $k = h/m$ at $(0, -a, z)$. The vortex line is assumed to be at rest with respect to the surface[20] to simulate a probable condition for the ring just after creation near a bubble. The appropriate boundary condition is that the pressure be constant at all points on the surface. We assume that the surface is only slightly deformed and that the velocity of the fluid is given by the sum of the velocity fields of the vortex line and its image. The velocity at the surface is tangential and given by

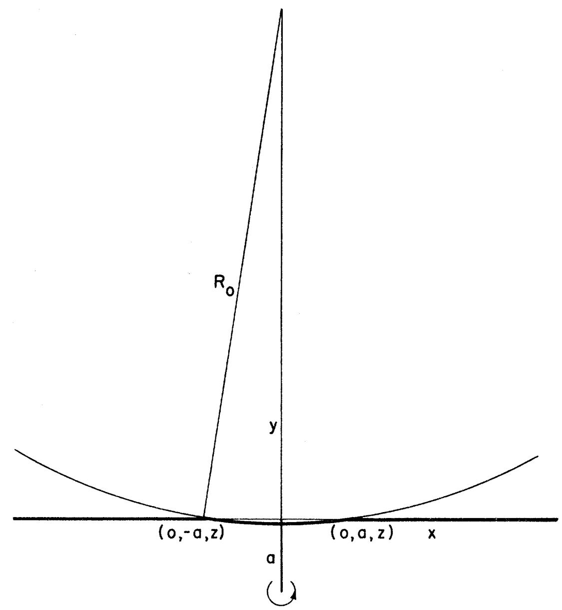

FIG. 1. Coordinate system used in calculating the surface deformation energy caused by a vortex line located at $(0, -a, z)$. The liquid surface is outlined by the heavy line and lies in the $y = 0$ plane except for a depression near the origin. The z axis extends out of the page.

$$v(x) = -ka/\pi(x^2 + a^2) . \tag{6}$$

The Bernoulli pressure depresses the surface near the vortex line and is equal to

$$p(x) = -\rho k^2 a^2/2\pi^2(x^2 + a^2)^2 . \tag{7}$$

This pressure is balanced by the surface-tension pressure which is equal to the product of the surface-tension constant σ and the curvature of the surface. The Bernoulli pressure is small for $|x| > a$. We approximate the shape of the surface by an arc of radius R_0 which intersects the surface at $x = \pm a$. We chose R_0 to give the correct surface-tension pressure at $x = 0$:

$$R_0^{-1} = \rho k^2/2\pi^2 a^2 \sigma . \tag{8}$$

We limit ourselves to the case $R_0/a \gg 1$ which implies $a \gg 2$ Å for liquid helium. The surface deformation energy is equal to the surface-tension constant times the change in area of the surface. Within the above approximation the surface deformation energy per unit length of vortex line is

$$E_d/L = \tfrac{1}{3}\sigma R_0 (a/R_0)^3 = \rho^2 k^4/12\sigma\pi^4 a \ . \tag{9}$$

For a vortex line segment of length equal to the circumference of a 15 Å bubble we obtain

$$E_d \simeq 3/a \text{ meV} ,$$

where a is measured in angstroms.

The surface of a bubble is much less compressible than a free surface, but for small a the energy associated with the deformation of the bubble may be a fraction of the kinetic energy of the ion at its critical velocity which is approximately 4 meV. This term should decrease as the external pressure is increased and the bubble becomes more incompressible. The critical velocity for the negative ion now becomes

$$V_c = (E + E_d)/P + P/2m^* \ . \tag{10}$$

If the surface deformation energy is to first order proportional to the radius of the ion, then the critical velocity remains inversely proportional to the radius. The effect of the condensation of He^3 atoms on the surface of the ion in this creation process is not clear.

This model of the condensation of He^3 atoms on the surface of negative ions can be tested by further studies of the pressure, temperature, and concentration dependence of V_c. A measurement of the critical velocity in the presence of normal fluid flow might be useful in determining the importance of the role of the normal fluid in the creation process. Measurements of surface tension at very low temperatures with small concentrations of He^3 should also determine whether the He^3 atoms condense on the surface of bulk He.[4]

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