Nature of Exotic Negative Carriers in Superfluid ^{4}He

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The mysterious "fast" and "exotic" negative carriers in liquid ^{4}He are interpreted as multielectron bubbles. The fast carrier is shown, by a detailed calculation of mobility, to be a two-electron bubble with a radius of approximately 30 Å. A crucial role is played by the unusual kinematics of rotons, which have not previously been adequately treated. The other exotic carriers are multielectron bubbles with higher charges.

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The "normal" negative carrier in liquid helium is well established as a single electron in a bubble state, with the electron wave function concentrated in a spherical region, about 16 Å in radius, essentially free of He atoms.[1] Another ("fast") negative carrier, first reported in 1969,[2] was confirmed and studied in 1971[3,4] in experiments which reported the existence of still other ("exotic") negatively charged entities. These observations have more recently been confirmed,[5] but the nature of the objects has remained obscure. References 3 and 4 discuss, and find reasons to exclude, various possible models.

One obvious possibility is that these are many-electron bubbles. The two-electron bubble is energetically unstable against decay into two one-electron bubbles, as is discussed below. In addition, if the cross section for roton scattering were to scale with the square of the radius, the mobility of the two-electron bubble (calculated radius R approximately 30 Å) would be less than that of the normal carrier. However, the fast carrier has a mobility approximately 6 times *larger* than the normal one. We suggest, nonetheless, that the fast carrier is a two-electron bubble, which is stable against *small* deformations, and that the exotic carriers with intermediate mobilities are many-electron bubbles with higher charges. We find that this interpretation gives an accurate account of the observed mobilities, provided that proper attention is paid to the unusual kinematics of rotons.

This work is concerned with carrier mobilities in the temperature range near 1 K, where the elementary excitations primarily responsible for the frictional drag come from the roton region of the elementary excitation spectrum. At these temperatures, furthermore, the roton mean free path is large compared with the dimensions of the carriers considered. The known carriers also have large effective masses. Under these conditions the mobilities can be calculated with the theory of Baym, Barrera, and Pethick.[6] Since the carriers may possess Z electrons, we evaluate the mobility μ and the friction coefficient $\gamma=F/v$ from the expression

$$\gamma=Ze/\mu=-(6\pi^2\hbar^3)^{-1}\int_0^\infty p^4[\partial n(p)/\partial\epsilon]\,|d\epsilon/dp|\,\sigma_m(p)dp, \tag{1}$$

where the momentum-transfer cross section $\sigma_m(p)$ can be evaluated with use of

$$\sigma_m(p)=\int p^{-2}[\mathbf{p}\cdot(\mathbf{p}-\mathbf{p}')](d\sigma/d\Omega)d\Omega. \tag{2}$$

In this expression, $\mathbf{p}$ and $\mathbf{p}'$ are initial and final roton momenta, and $n(p)$ is the occupation of state p by a roton

$$n(p)\{\exp[\epsilon(p)/\tau-1]\}^{-1}\cong\exp[-\epsilon(p)/\tau].$$

The roton energy $\epsilon(p)$ is customarily written in the form

$$\epsilon(p)=(p-p_0)^2/2\mu_p+\Delta,\quad \tau=k_BT\ll\Delta=8.68\text{ K}. \tag{3}$$

The quantity $p^{-2}[\mathbf{p}\cdot(\mathbf{p}-\mathbf{p}')]$ reduces to $1-\cos\theta_{pp'}$ and is between 0 and 2, for the usual elastic scattering, where $p=p'$.

Rotons, however, have two ("conjugate") values of p, here called p and $\tilde{p}$, for a given energy. If we use the dispersion law given above, then $\tilde{p}=2p_0-p$. We will usually prefer the rationalized expression

$$\epsilon(p)=[(p^2-p_0^2)^2/(8p_0^2\mu_p)]+\Delta, \tag{4}$$

which differs fractionally from (3) by $(p-p_0)/p_0$, a small quantity in the temperature range of concern.[7] With the rationalized expression (4), conjugate roton momenta are related by $\tilde{p}^2=2p_0^2-p^2$. Figure 1 shows the situation for elastic reflection of rotons from a plane surface. Rotons with $p>p_0$ are denominated "ordinary" or "O," and those with $p<p_0$ "extraordinary" or "E." The group velocity, $\mathbf{v}=\nabla_p\epsilon$ is parallel to $\mathbf{p}$ for ordinary rotons, but *antiparallel* for extraordinary.[8] A pro-

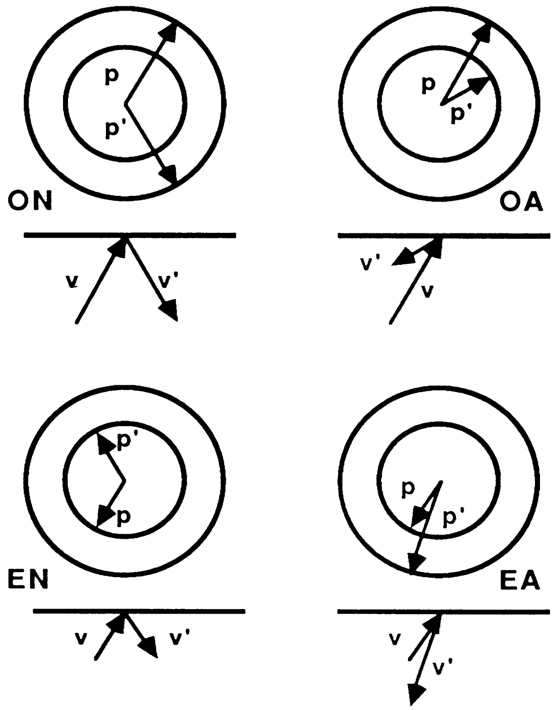

FIG. 1. Roton reflection from a plane surface. The concentric circles represent the two sheets (radii p and $\tilde{p}$) of a surface of constant energy in momentum space. In the space diagram, the roton is incident from below and reflected downward, with initial and final velocities shown. The four drawings illustrate normal (N) and anomalous (A) reflection of ordinary (O) and extraordinary (E) rotons.

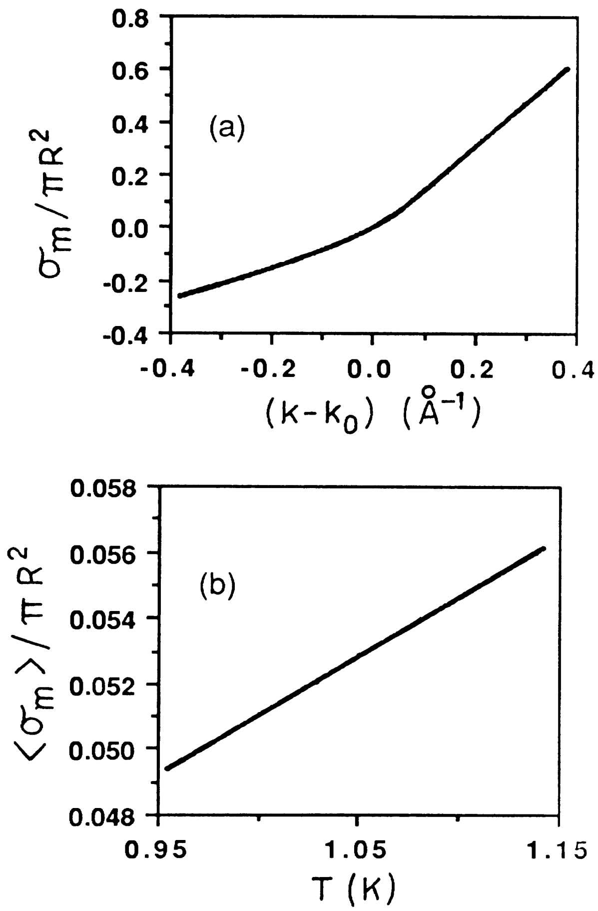

FIG. 2. (a) Roton momentum-transfer cross section as a function of wave number. (b) Averaged roton momentum-transfer cross section plotted against temperature.

cess is "normal" (N) if $p'=p$, and "anomalous" (A) if $p'=\tilde{p}\neq p$. For these elastic processes $\epsilon=\epsilon'$ and $p_\parallel=p'_\parallel$. The factor $p^{-2}[\mathbf{p}\cdot(\mathbf{p}-\mathbf{p}')]$, unlike $(1-\cos\theta_{pp'})$, can be less than zero, and is so for anomalous scattering of extraordinary rotons (EA scattering).

A given incident roton can, in general, undergo the two types of process (OA reflection is only allowed over a range of angles of incidence). The branching ratios (flux fractions), Γ_N and Γ_A, are the probabilities of the two processes. In a slowly varying force field, that is, in the classical limit, only one process will occur.[9] A quantum-mechanical argument determines the branching ratios: The function $\epsilon(p)$ is considered a Hamiltonian, with the substitution $\mathbf{p}\rightarrow(\hbar/i)\nabla$ for the passage to quantum mechanics. At this stage the preferred form of $\epsilon(p)$ is Eq. (4), which produces a fourth-order equation

$$\{[(-\hbar^2\nabla^2-p_0^2)^2/(8p_0^2\mu_p)]+\Delta\}\Psi=\epsilon\Psi. \tag{5}$$

In one dimension, this has four solutions, $\exp(\pm i\tilde{p}z/\hbar)$ and $\exp(\pm i\tilde{p}z/\hbar)$ for a given energy. A region in which the roton cannot propagate is represented by our letting $\Delta\rightarrow\infty$. Examination of this limiting process shows that Ψ and its first normal derivative must vanish at a hard wall, and yields a solution

$$\Psi=\exp\left(\frac{ipz}{\hbar}\right)+\alpha_N\exp\left(\frac{-ipz}{\hbar}\right)+\alpha_A\exp\left(\frac{i\tilde{p}z}{\hbar}\right),$$

whence

$$\Gamma_N=|\alpha_N|^2=[(\tilde{p}_z-p_z)/(\tilde{p}_z+p_z)]^2, \tag{6}$$

$$\Gamma_A=(\tilde{p}_z/p_z)|\alpha_A|^2=4p_z\tilde{p}_z/(\tilde{p}_z+p_z)^2. \tag{7}$$

We next consider the scattering from a hard sphere. We treat the problem classically, since $kR\cong 60$, expressing all quantities in terms of the impact parameter b, and integrating from 0 to R. The branching ratio Γ_N vanishes at the roton minimum, and the factor $[\mathbf{p}\cdot(\mathbf{p}-\mathbf{p}')]$ changes sign there for E rotons, so that the momentum-transfer cross section[10] has the unusual appearance[11] shown in Fig. 2(a). It can be expressed in closed form as

$$(\sigma_m/\pi R^2)_{\rm O} = \tfrac{1}{3}(2\rho^4 - 4\rho^2 - 1) - \tfrac{1}{2}\rho(\rho^2+1) + 4(\rho^2+\rho+1)/[3(\rho+1)] + \tfrac{1}{4}(\rho^2-1)^2\ln[(\rho+1)/(\rho-1)]$$

for $p > p_0$, and

$$(\sigma_m/\pi R^2)_{\rm E} = -1 - \tfrac{1}{2}\rho(\rho^2-1) + \tfrac{1}{3}\rho + 4/[3(\rho+1)] + \tfrac{1}{4}(\rho^2-1)^2\ln[(\rho+1)/(\rho-1)]$$

for $p < p_0$. In these expressions ρ denotes $\tilde{p}/p$. Inserting these cross sections in (1), we perform the integral over the roton thermal distribution numerically, writing

$$\langle\sigma_m\rangle \equiv \eta^{-1}\int_{-\infty}^{\infty}\sigma_m \exp(-\zeta^2/\eta)\,|\zeta|\,d\zeta, \tag{8}$$

with $\zeta \equiv (p-p_0)/p_0$ and $\eta \equiv (2\mu_\rho k_{\rm B}T/p_0^2)$, $k_0 = p_0/\hbar = 1.92$ Å^{-1}, and $\mu_\rho = 0.16M_{\rm He}$. Figure 2(b) shows a plot of this effective cross section as a function of temperature. There is considerable cancellation between ordinary and extraordinary rotons, leading to a cross section small compared with geometrical and slowly increasing with temperature. The line shown is $\langle\sigma_m\rangle = 0.051T^{0.69}\pi R^2$. The friction coefficient or mobility can then be calculated from

$$\gamma = Ze/\mu = \hbar k_0^4/3\pi^2 \exp(-\Delta/\tau)\langle\sigma_m\rangle = \hbar k_0^4/3\pi^2\exp(-\Delta/\tau)0.051T^{0.69}\pi R^2. \tag{9}$$

We now require predictions for R from the bubble model.

The one-electron bubble has been well studied both theoretically and experimentally. The electron wave function is confined to a spherical cavity whose equilibrium radius at $T=0$ ($R_0^{(1e)} \cong 16$ Å) is determined by the minimization of the sum of electron energy and surface energy. The total energy is approximately 0.19 eV.

The two-electron system has been analyzed theoretically by Dexter and Fowler.[12] Its equilibrium radius is again determined by the minimization of the sum of electron and surface energy, but the problem is significantly complicated by Coulomb repulsion. A variational calculation leads to a reasonably reliable estimate of the energy, which is found to be 0.84 eV, considerably higher than that of two one-electron bubbles. Thus the two-electron bubble is not absolutely stable against decay into two one-electron bubbles, but it may well be stable against small deformations. The energy is minimized for $R_0^{(2e)} \cong 60a_0 \cong 32$ Å; the same calculation somewhat overestimates $R_0^{(1e)}$, obtaining a value of 17.5 Å. Bubbles in equilibrium under surface and Coulomb energies alone are marginally stable.[13,14] Electron kinetic energy is small but not negligible for few-electron bubbles, and should have a stabilizing influence. We will assume them to be sufficiently stable, and calculate mobilities using the theory of the previous section.

Figure 3 shows experimental mobilities from Fig. 17 of Ref. 4 for the fast carrier. The line through the points is from Eq. (9) with $R^2/Z = 435$ Å^2, i.e., a two-electron bubble with $R = 29.5$ Å. The mobility calculated with the theoretical value, $R_0^{(2e)} = 32$ Å is also shown. This agreement is remarkable, considering that no adjustable parameters have been used. The temperature dependence of $\langle\sigma_m\rangle$ leads to an effective activation energy near $T = 1$ K of

$$-d(\ln\mu)/d(1/T) = 8.68\ \text{K} + 0.69(1\ \text{K}) = 9.4\ \text{K},$$

giving very good agreement with the observed slope.

We can estimate mobilities of carriers with higher charge. According to (9), $\mu \propto Z/R^2$. We can crudely estimate the variation of radius R with charge Z for many-electron bubbles by including only Coulomb and surface energy, ignoring kinetic energy (and shell structure), and variation in charge distribution with Z. Such a gross approximation predicts $R \propto [Z(Z-1)]^{1/3}$ or $\mu \propto Z/[Z(Z-1)]^{2/3}$. The first three exotic carriers have mobilities lower than the fast carrier by factors of approximately 0.67, 0.62, and 0.59, respectively.[4] Our crude approximation predicts mobilities for $Z = 3$, 4, and 5 relative to $Z = 2$ of 0.72, 0.61, and 0.54; we regard the agreement as very satisfactory, considering that $Z = 3$ represents opening of the first p orbital and that $Z = 5$ is a half-filled p shell, which is presumably spherical again.

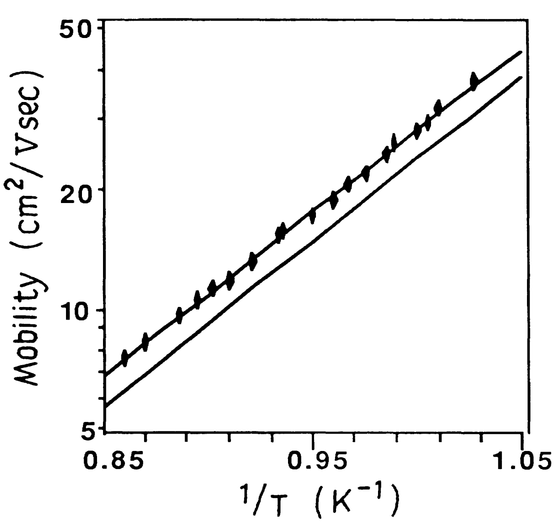

FIG. 3. Mobility vs temperature. The data points are for the "fast" carrier from Ref. 4. The upper line is calculated from Eq. (9) with $R = 29.5$ Å; the lower line is similarly calculated with the theoretical value $R_0^{(2e)} = 32$ Å.

The present theory does not give a good account of the mobilities of the well-known "normal" negative and positive carriers. This model predicts an effective cross section approximately one-twentieth geometrical. The positive carrier, however, has a radius about 6 Å, and a roton cross section approximately twice geometrical. The normal negative (one-electron bubble) has a radius of approximately 16 Å, and a cross section approximately one-half geometrical. The source of the discrepancy may be the relatively small size of these objects. On the one hand, semiclassical theory is less valid; on the other, the liquid helium outside these objects is significantly perturbed in the strong electric field surrounding each. Much of the scattering may not occur at the "hard-sphere" radius, but in the surrounding force field. If the main effect is the decrease in Δ associated with the high pressure near the carrier, there will be no anomalous scattering in the classical approximation. This will tend to increase the cross section.

The model provides no natural explanation for one of the most striking properties of the fast carrier, namely the fact that it appears to reach the Landau critical velocity (60 m/sec) without creating vortex rings. Nonetheless, the evidence presented here seems convincing. The multielectron bubbles are expected to be rather "soft" against quadrupole distortions. They may thus have much lower d-wave resonant frequencies, and much stronger phonon scattering than the one-electron counterpart.[6] The conditions under which these novel negative carriers are produced[3-5] apparently involve charging of the free surface. Thus, they resemble those under which the giant negative bubbles[15] occur.

Finally, it would be interesting to study experimentally the various reflections illustrated in Fig. 1. When elastic reflection occurs at a crystalline surface, $p_{\parallel}$ can change by a reciprocal-lattice vector. Rotons might be a useful surface probe.

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