New Approach to the Observation of the Condensate Fraction in Superfluid Helium-4

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We present a phenomenological analysis of an experiment to observe the condensate fraction of helium-4 by firing low-energy pulses of helium atoms at suspended droplets of the superfluid and observing the resulting emission of helium atoms from the fluid. Emission occurs through a conventional process in which rotons are produced and propagate to the other side of the droplet, and through a second process depending on the existence of the condensate. If A is the area of the incoming beam, the cross section scales with A^2 (first process) and A^4 (second process), respectively, for small A. Aspects of a full many body calculation and the performance of such an experiment are briefly discussed.

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Superfluid helium-4 is the only boson superfluid known to occur in nature. It is interesting that though the basic origin of the superfluidity, in a boson condensation, has been known for many years [1] experimental and theoretical details concerning the microscopic nature of the order parameter associated with the transition to the superfluid state remain sketchy. Formal relationships connecting the microscopic theory with the two fluid hydrodynamics exist [2–4] but no detailed theory relates microscopic wave functions for the fluid [5] directly to the hydrodynamic properties of the superfluid [6]. A deeper understanding of the microscopic nature of superfluid helium-4 would have fundamental interest and could lead to useful insights in such areas as possible superfluidity in planetary interiors and the search for other boson superfluids.

Here we propose a new kind of experiment to obtain information about the condensate fraction in superfluid helium four by studying elastic scattering of helium atoms from a freely floating macroscopic sphere of the fluid. If successful, the experiment would be important because direct experimental study of the condensate fraction has proved extremely elusive. Only neutron scattering experiments give direct information [7,8], and interpretation of it has proved difficult. Indeed it is remarkable that Ref. [8] concludes its review of many years of experimental work with the statement that "a direct observation of the condensate fraction, has not come to pass.... It is unlikely that this goal will ever be reached by deep inelastic neutron scattering experiments."

Our basic idea is that in a configuration of liquid helium confined by two parallel free surfaces (as in a large suspended droplet in a microgravity environment), it will be possible to do an experiment (analogous to a Josephson tunneling experiment in some respects) in order to study the condensate. We envision sending a pulsed beam of gaseous helium atoms at one side of the superfluid helium-4 and detecting helium atoms emerging elastically and immediately thereafter from the other side. The experiment we propose would be difficult without two or more widely separated free surfaces. In more convoluted geometries (such as, for example, two horizontal parallel surfaces connected by a curved tube filled with superfluid) controls associated with the existence of signals from the competing process of roton creation and annihilation would be absent. Further, in many such possible geometries, there is a strong coupling to a macroscopic thermal environment, possibly leading to poorly understood dissipative processes. One could in principle try to look at backscattered helium atoms but it is not possible to distinguish the process of interest from other simple backscattering processes having nothing to do with the condensate. In the proposed configuration, backscattering is not detected at all. Further, by making the beam size A less than the total geometrical area of the helium sphere, we ensure that no forwardly moving helium from the incident beam interferes with the measurement.

The experiment would obtain information about the condensate fraction from the intensity of helium atoms emerging elastically and promptly in the forward direction from the superfluid helium after it is probed with the pulsed beam. In particular, the dependence of this intensity on the area of the quantum mechanically coherent incoming pulse will be anomalous, when the process leading to emission depends directly on the presence of the condensate. To describe this idea, suppose for simplicity that the surfaces are perfectly parallel so that the problem looks like the tunneling of a helium particle through a slab of liquid helium. (See Fig. 1. We have not made a detailed study of the focusing effects of a spherical droplet.) One can understand how this experiment can directly access the condensate fraction of the liquid helium by referring to Yang's original association [9] of the condensate fraction with off diagonal long range order:

$$n_0 \equiv N_0/N = \lim_{|\mathbf{r}_1-\mathbf{r}_1'|\to\infty} N\rho^{-1}\int d\mathbf{r}_2\cdots\int d\mathbf{r}_N\,\Psi^*(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N)\Psi(\mathbf{r}_1',\mathbf{r}_2,\ldots,\mathbf{r}_N)\,, \quad (1)$$

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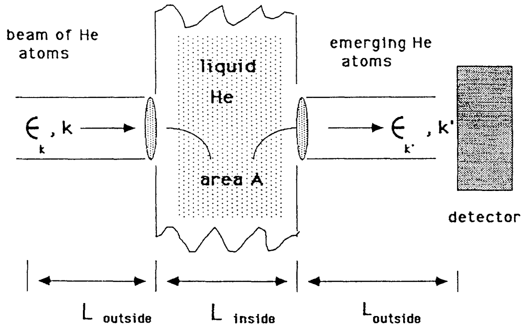

FIG. 1. Sketch of essential features of the proposed experiment.

where N_0 is the number of particles in the condensate of superfluid containing N particles, $\Psi(\mathbf{r}_1, \ldots, \mathbf{r}_N)$ is the N-body wave function of the fluid, and $\rho = N/V$, where V is the fluid volume. From this fundamental definition, one sees that, ideally, one would want to measure the amplitude for adding a particle to the fluid at one point ($\mathbf{r}_1$) and removing it from a far distant point $\mathbf{r}_1'$ in order to measure the condensate. The proposed experiment comes very close to doing precisely this. Unlike the traditional neutron scattering experiment, the experiment will probe the fluid at the low energies and momentum characteristic of the condensate fraction itself and is therefore free of the corrections to the impulse approximation (not required here) which plagued the neutron scattering experiments [8].

The full interpretation of the experiment requires accurate assessment of the local amplitudes for absorbing an incident particle into the fluid at one side and for emitting a particle at the other side. This requires a theoretical calculation which we have only done quite crudely. However, we stress that our analysis shows that the *existence* of the condensate can be verified in the proposed experiment even without such a calculation. There may be competition between the process of interest and another, classical, process in which a sound wave (here a phonon roton) is produced at one side, propagates across the fluid, and causes reemission of a helium atom at the other side. We will show that this process scales differently from the process of interest with the cross-sectional area of the incoming pulse when that area is small, and thus can, in principle, be distinguished from it.

To calculate the amplitude of the process of interest we need to know the many body wave function describing the $(N+1)$-body system consisting of the droplet and an incoming or outgoing particle in the vapor. For a preliminary discussion, we will describe the coupling of the incoming and outgoing particles to the fluid by a kind of transfer Hamiltonian [10], somewhat as in early analysis of electronic tunneling experiments [11]:

$$H' = \sum_{\mathbf{k}} (T_{\mathbf{k}}^L b_{\mathbf{k},L} a_0^\dagger + T_{\mathbf{k}}^R b_{\mathbf{k},R}^\dagger a_0 + \text{H.c.}) . \quad (2)$$

$b_{\mathbf{k},L,R}^\dagger$ create particles coming in from the left and leaving to the right, respectively, and $a_0^\dagger$ create particles in the fluid condensate. (Momentum is conserved in the implied processes through distribution of the momentum of the incoming particle among a macroscopic number of particles in the fluid.) The process of interest is one in which the condensate virtually absorbs a particle which is then coherently reemitted at the other side of the sample. In lowest order, this will be of second order in H' and the Fermi "golden rule" obviously gives a rate of

$$\frac{2\pi}{\hbar} N_0^2 \sum_{\mathbf{k}'} \frac{|T_{\mathbf{k}}^L T_{\mathbf{k}'}^R|^2}{(\epsilon_{\mathbf{k}} - E_{N+1} + E_N)^2} \delta(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}'}) , \quad (3)$$

where E_N is the fluid ground state energy when the fluid has N particles and N_0 is the number of particles in the condensate (assumed $\gg 1$). The factors T are expected to scale as

$$T = t \left(\frac{\lambda A}{V} \right)^{1/2} \left(\frac{\lambda A}{L_{\text{outside}} A} \right)^{1/2} = t\lambda \left(\frac{A}{V L_{\text{outside}}} \right)^{1/2} \quad (4)$$

in which the A is the cross-sectional area of the incoming beam of helium atoms, λ is a length of the order of the width of the interface, V is the entire volume of the liquid helium sample, and L_{outside} is the length normal to the interfaces of the cylindrical volume in which the wave functions of the incoming and outgoing beams are normalized. t is a matrix element with the dimensions of energy which will depend on the microscopic physics of the interface and also on geometric factors associated with collection efficiency. t is on the scale of atomic energies per particle. The rate can be expressed as a cross section of the floating helium droplet for the process of absorption and reemission as described:

$$\sigma = \frac{2\pi m}{\hbar^2 k} \frac{N_0^2 A^4}{V^2} \int \frac{d\mathbf{k}'}{(2\pi)^3} \frac{\lambda^4 |t_{\mathbf{k}}^L t_{\mathbf{k}}^R|^2}{(\epsilon_{\mathbf{k}} + |\mu|)^2} \delta(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}'}) . \quad (5)$$

The interesting feature is the N_0^2 whose origin is not hard to trace and which leads to a very unusual dependence on the beam area A in lowest order in H'. Expressing the result in terms of the atomic volume density ρ of the liquid helium and the condensate fraction $n_0 = N_0/N$ one finds

$$\sigma = \frac{2\pi m}{\hbar^2 k} n_0^2 \rho^2 A^4 \int \frac{d\mathbf{k}'}{(2\pi)^3} \frac{\lambda^4 |t_{\mathbf{k}}^L t_{\mathbf{k}}^R|^2}{(\epsilon_{\mathbf{k}} + |\mu|)^2} \delta(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}'}) . \quad (6)$$

The striking dependence on A arises because N_0^2 increases with the square of the volume of the liquid helium.

It is instructive to compare the result with the corresponding one for a process in which a roton is virtually excited in the intermediate state. The result is

$$\sigma' = \frac{2\pi m}{\hbar^2 k} A \frac{A}{L_{\text{inside}}^2} V \int\int \frac{d\mathbf{q}}{(2\pi)^3} \frac{d\mathbf{k}'}{(2\pi)^3} \frac{\lambda^4 |t_{\mathbf{k},\mathbf{q}}^L t_{\mathbf{k}',\mathbf{q}}^R|^2}{(\epsilon_{\mathbf{k}} - \hbar\omega_{\mathbf{q}} + |\mu|)^2} \times \delta(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}'}) . \quad (7)$$

Here the cross section is proportional to A^2. A second factor of A arises because incident flux here is proportional to $1/A$ (because the pulse is assumed to be smaller in cross section than the target, unlike the standard scattering calculation). L_{inside} is the length of a cylinder inside the fluid as shown in Fig. 1. (We have neglected diffraction of the roton beam.) The sum on roton wave vectors $\mathbf{q}$ will be strongly constrained by the requirement that the roton be moving almost exactly normal to the interface. The integral on $\mathbf{q}$ involves the product of the phonon-roton density of states at $\omega_{\mathbf{q}}$ and the factor $1/(\epsilon_{\mathbf{k}}-\hbar\omega_{\mathbf{q}}+|\mu|)^2$ which will be small over most of the integration region as long as $\epsilon_{\mathbf{k}} < -|\mu|+\hbar\omega_{\text{roton}}$ or $\epsilon_{\mathbf{k}} > -|\mu|+\hbar\omega_{\text{maxon}}$. (The integral on $\mathbf{q}$ as written contains a singularity at $\omega_{\mathbf{q}}=\epsilon_{\mathbf{k}}+|\mu|$. This is analogous to the resonant Raman effect and can be handled similarly by including quasiparticle lifetimes.) The constraint $\epsilon_{\mathbf{k}} < -|\mu|+\hbar\omega_{\text{roton}}$ or $\epsilon_{\mathbf{k}} > -|\mu|+\hbar\omega_{\text{maxon}}$ means quantitatively that we require $\epsilon_{\mathbf{k}} \lesssim 2$ K or $\epsilon_{\mathbf{k}} \gtrsim 7$ K for the energy of the incoming beam. The second range is probably better, but many other processes will begin to enter if the energy is not kept low. One way to distinguish the process involving the condensate [Eq. (6)] experimentally would be to look for the part of the signal (time correlated with the incident pulse) which scaled as A^4 instead of A^2, in this energy range in which the roton-maxon process should be weak. For the condensate mediated process, the prediction that the cross section scales as A^4 depends on the lowest order nature of the calculation just described.

To obtain a preliminary estimate of the matrix element we have made a calculation in which the incoming wave is described as a plane wave reflected from a step function distribution of helium fluid beginning at the interface and the condensate wave function is described as the square root of the product of the total density of the fluid and the local condensate fraction as calculated for a helium fluid surface by Krotscheck [12]. Using this in Eq. (6) suggests that the lowest order calculation is only adequate if the radius of the incoming pulse is much less than a tenth of a micron. For larger areas, the area dependence will change, and presumably become weaker. The same estimates suggest that the condensate mediated process will dominate when the geometrical area of the incoming beam is more than about a tenth of a micron in radius.

We have not made a detailed calculation of the time dependence of pulses emitted due to these processes after an incoming wave packet of helium atoms encounters the liquid. Simple uncertainty principle considerations suggest that the time delays are of order $\hbar/\Delta E$ where ΔE is the energy denominator in either Eq. (6) or (7). On this basis one sees that the time delays for the two processes are comparable and of the order of a picosecond. The resonant process associated with phonon or roton creation occurs when the energy denominator in Eq. (7) is zero. Then the time delay can be estimated as L_{inside}/v_s where v_s is the velocity of the created quasiparticle. If L_{inside} ≈ 1 mm then this is a few microseconds or more and one could hope to resolve it by existing time of flight techniques. To obtain a resonant denominator requires incoming energies of 5 K or less.

The coherent effects on which the effects described depend require a helium atom incident normal to the surface. The tolerance with which one must achieve this alignment of the incoming wave vector in the normal direction may be roughly estimated as (in radians) $1/k\sqrt{A}$, where k is the magnitude of the incoming wave vector. If $\sqrt{A}$ is of the order of a tenth of a micron and the wavelength of the incoming beam is a few angstroms then the tolerance is around a percent of 4π.

Coherent helium atom beams are used routinely for surface scattering experiments [13], but in the proposed experiment we require beams with a much lower translational energy than is usually used in those experiments. Simply reducing the temperature of the beam source will not necessarily work, because of the increasing likelihood of forming clusters. Recent experiments on helium atom beams from small nozzles [14] show that, for example, the beam from a nozzle 5 μm in diameter with a source temperature of 5.5 K and pressure of 8 bars provides a mixture of clusters and single atoms. Fortunately, the atoms can be distinguished by their higher velocity. Under these conditions the atom wave velocity is centered at 235 m/s with a width of about 16%, and the average wavelength of the atoms is about 0.42 nm. If such a source were placed 1 m from a suspended droplet, the first diffraction zone of the waves would be 0.2 mm in diameter and the use of masks would be feasible to control the area A. If such masks can be made in the form of Fresnel zone plates, we may consider the possibility of refocusing the beam to a desired, controllable size. As the temperature of the gas in the stagnation chamber is lowered towards its boiling point, the intensity of the atom beam decreases as more clusters are formed. In an ideal supersonic expansion the beam will have a kinetic energy of $\frac{5}{2}k_BT$, where T is the temperature of the stagnation chamber and k_B is Boltzmann's constant. This appears to put a lower limit of about 10 K on the kinetic energy of a helium atom beam from such a stationary source.

To avoid extreme levels of gas loading in the chamber containing the droplet, the use of a pulsed beam is desirable and techniques for obtaining pulsed beams are available [15]. The pulsed valves are typically sealed using an elastomer, however, and such a seal will not function at cryogenic temperatures. Valves using metal-to-metal or metal-to-sapphire seals have been built, but they are much harder to prepare and have not been tried at low temperatures. It is likely that a cryogenic valve can be developed using such a seal. A recent report [16] describes a very interesting technique for forming very slow (kinetic energy ≈ 1.5 K) pulsed beams of helium, but the cluster composition of the beam has not been measured directly. Matter wave interference effects have been observed for coherent helium atoms at higher temperatures [17].

The detection of helium atoms in the proposed experiment requires a fast, low noise atom detector. In order to make the mean free path of helium in the scattering chamber large enough, the chamber must be kept at about 0.3 K or less. This low temperature chamber would be an ideal environment for a bolometer [16,18, 19], so that is probably the best choice for the atom detector. The use of bolometers for detecting low-energy helium atoms is well known [20] and has recently been reported [16] at the energies needed for the experiment proposed here.

To get a number for n_0 from the proposed experiment we would need values for the matrix elements t. For this one must find the many body ground state wave functions of liquid helium with $N+1$ particles and the appropriate boundary conditions. To characterize the boundary conditions in a convenient way, we define the functions

$$f_j(\mathbf{r}_1,\ldots,\mathbf{r}_{N+1})=\frac{\Psi_{N+1}(\mathbf{r}_1,\ldots,\mathbf{r}_{N+1})}{\Psi_N(\mathbf{r}_1,\ldots,\mathbf{r}_{j-1},\mathbf{r}_{j+1},\ldots,\mathbf{r}_{N+1})}\,. \tag{8}$$

Here $\Psi_{N+1}(\mathbf{r}_1,\ldots,\mathbf{r}_{N+1})$ is the many body ground state wave function for the $(N+1)$-body system with boundary conditions such that $f_j\rightarrow\mathcal{A}e^{i\mathbf{k}\cdot\mathbf{r}_j}+\mathcal{B}e^{-i\mathbf{k}\cdot\mathbf{r}_j}$ as $\mathbf{r}_j$ gets far from the liquid to the left while the other coordinates stay in the liquid while $f_j\rightarrow e^{i\mathbf{k}\cdot\mathbf{r}_j}$ as $\mathbf{r}_j$ gets far from the liquid to the right while the other coordinates stay in the liquid. The coefficient $\mathcal{A}$ can be interpreted as the reciprocal of the transmission coefficient characterizing the process by which a helium atom appears on the right as a consequence of an incident particle impinging on the fluid from the left with momentum $\hbar\mathbf{k}$. The rate of such transmission is then $\hbar|\mathbf{k}|/mL_{\text{outside}}\mathcal{A}^2$ and the cross section is $\sigma=A/\mathcal{A}^2$. To calculate the wave function one can adapt the Feenberg-Jastrow Euler-Lagrange method which has been a highly successful theory of the bulk fluid ground state [21–23], including the structure of a free surface [24–26]. The adaptation of this theory to the state Ψ_{N+1} is relatively straightforward.

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