Surface Region of Superfluid Helium as an Inhomogeneous Bose-Condensed Gas

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(Received 17 May 1995)

We present arguments that the low density surface region of self-bounded superfluid ^{4}He systems is an inhomogeneous dilute Bose gas, with almost all of the atoms occupying the same single-particle state at $T = 0$. Numerical evidence for this complete Bose-Einstein condensation was first given by the many-body variational calculations of ^{4}He droplets by Lewart, Pandharipande, and Pieper in 1988 [Phys. Rev. B **37**, 4950 (1988)]. We show that the low density surface region can be treated rigorously using a generalized Gross-Pitaevskii equation for the Bose order parameter.

PACS numbers: 67.40.Db, 05.30.Jp

In recent years, there has been a major effort to produce atomiclike gases in a Bose-condensed phase, using excitons, spin-polarized hydrogen, and laser-trapped alkali atoms (for reviews of this work, see Refs. [1,2]). In such systems, the fraction of atoms in the lowest energy single-particle state could approach 100% at $T = 0$. This unique phase of matter was first studied by Einstein and London and would exhibit many unusual properties [2], including superfluidity. In contrast, in bulk superfluid ^{4}He the condensate fraction is only about 10% at $T = 0$ [1,3]. Thus the ground state wave function does not have the simplicity of a gas with complete Bose-Einstein condensation (BEC).

In the present Letter, we argue that such a dilute Bose-condensed gas is in fact already present in the surface region of superfluid ^{4}He, where the density becomes very small. This striking phenomenon has apparently been overlooked in the extensive theoretical literature on surfaces, films, and droplets of superfluid ^{4}He (for references, see [4]). Moreover, in this low density surface region, where one has almost 100% BEC into a single state, one can do microscopic calculations of the order parameter using the standard field-theoretic description of an inhomogeneous Bose-condensed fluid [5,6]. Our discussion is limited to ground-state properties ($T = 0$) but we make a few remarks about finite temperature at the end.

Striking confirmation of the preceding physical argument is available in the results of Lewart, Pandharipande, and Pieper (LPP) [7], who studied ^{4}He droplets (with up to 240 atoms) using a variational ground-state wave-function approach [8]. These authors calculated the single-particle "natural orbitals" $\psi_i(\mathbf{r})$ which diagonalize the single-particle density matrix, giving

$$\rho_1(\mathbf{r},\mathbf{r}') = \sum_i n_i \psi_i^*(\mathbf{r})\psi_i(\mathbf{r}'), \tag{1}$$

where n_i is the occupation probability of the state $\psi_i(\mathbf{r})$. The local density is given by $\rho(\mathbf{r}) = \rho_1(\mathbf{r},\mathbf{r})$. By direct numerical calculation, LPP found that the nodeless $1s$ state $\psi_{1s}(\mathbf{r})$ had an occupation probability of $n_{1s} \simeq 25$, while for all the other states, $n_i < 0.5$. They identified (and justified) this $1s$ state as the Bose-condensed state, with an associated local condensate density being given by

$$\rho_c(\mathbf{r}) = n_{1s}|\psi_{1s}(\mathbf{r})|^2. \tag{2}$$

LPP evaluated the total local density $\rho(\mathbf{r})$ and the condensate local density $\rho_c(\mathbf{r})$ using (2). Their results for $N = 70$ atoms are replotted in Fig. 1. However, LPP did not explicitly discuss the *implications* of the most striking feature shown in Fig. 1, namely, that since in the surface region almost all the atoms at a given value of r are in the $\psi_{1s}(\mathbf{r})$ state, this surface region provides a physical realization of the long sought for dilute Bose-condensed gas [1,2].

An important aspect of the calculations in Ref. [7] is that near the center of the drop $\rho(\mathbf{r})$ and $\rho_c(\mathbf{r})$ are very close to the values in bulk superfluid ^{4}He, with $\rho_c(\mathbf{r}) \simeq 0.1\rho(\mathbf{r})$. This indicates that even droplets of $N = 70$ atoms are large enough to ensure that the surface region density profile will be very similar to that of a planar free surface of bulk superfluid ^{4}He. This is confirmed by more recent calculations on larger droplets [9]. In the case of planar (versus spherical) symmetry, the atoms will Bose condense into states with zero momentum parallel to the surface [10].

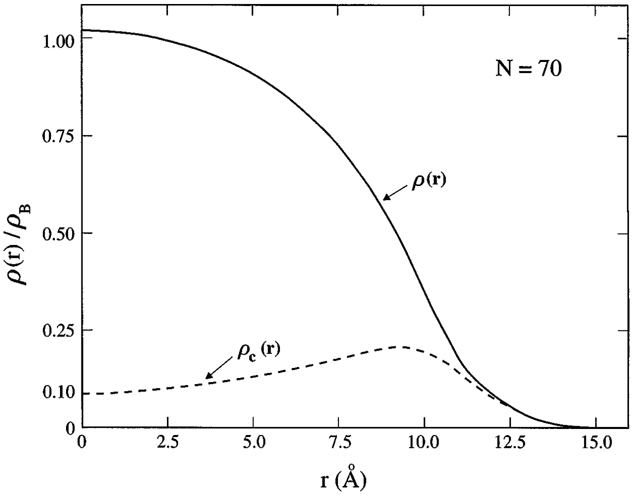

FIG. 1. The total density $\rho(r)$ and condensate density $\rho_c(r)$ as a function of the distance from the center of a ^{4}He droplet of 70 atoms, based on Fig. 5 of Ref. 7. For simplicity, we have smoothed out the small oscillations in $\rho(r)$.

Krotscheck [10] first found that the local condensate density $\rho_c(\mathbf{r})$ increased when the total density $\rho(\mathbf{r})$ decreased in liquid ^{4}He in contact with a wall. Campbell [11] has recently drawn attention to the results in Refs. [7–10] and discussed them in the context of the formal theory of off-diagonal long-range order (ODLRO) in liquid ^{4}He droplets. Apart from these papers, Bose condensation has been ignored in the literature on inhomogeneous Bose liquids at $T = 0$ (for references, see [4] and [12]). Moreover, even in Refs. [7–11] what is still missing is a physical interpretation of the numerical results for $\rho_c(\mathbf{r})$ at surfaces as evidence for (and relevance of) a low density Bose gas in which the condensate fraction can be 100%, and the important *implications* of this new picture. It is this aspect which the present Letter addresses.

The numerical results in Fig. 1 are consistent with the empirical formula given in Ref. [7],

$$\rho_c(r) = \rho(r)[1 - 0.68\rho(r)/\rho_B]^2, \tag{3}$$

where ρ_B is the bulk or saturation density of superfluid ^{4}He at $T = 0$. It is easy to verify using (3) that the local condensate *fraction* $\rho_c(r)/\rho(r)$ smoothly increases from 0.1 to unity as $\rho(r)$ goes from ρ_B to zero. One of the most interesting features shown by Fig. 1 is that $\rho_c(r)$ develops a maximum. Using (3), this occurs when $\rho(r) \simeq 0.5\rho_B$ and corresponds to $\rho_c(r) \simeq 0.22\rho_B$. Thus the results of Ref. [7] predict that in a certain region, the density of ^{4}He atoms which are Bose condensed can be *twice* as large as the bulk value $\rho_c \simeq 0.1\rho_B$. Of course, as $\rho(r)$ decreases to zero, so must $\rho_c(r)$. While the precise magnitude may depend on the approximate calculations used in [7], we believe that the *increase* in $\rho_c(r)$ as we go from the center of the ^{4}He drop is a real effect. As noted in Ref. [7], the initial increase in $\rho_c(r)$ shown in Fig. 1 for $\rho(r) < \rho_B$ is consistent with the calculated and observed decrease in ρ_c in bulk liquid ^{4}He under pressure [13]. Thus in superfluid ^{4}He, $\partial\rho_c(\rho)/\partial\rho \simeq -0.3$ for ρ close to ρ_B. In contrast, in a dilute hard sphere Bose gas, $\partial\rho_c(\rho)/\partial\rho \lesssim 1$ [5].

Independently of the numerical results in Refs. [7,9], it is easy to see that the general structure of the variational many-particle wave function which LPP and others [8,9,14–16] have used to treat the free surface of liquid ^{4}He already builds in the essential physics of a surface region which is completely Bose condensed into one single-particle state. In their simplest form, such wave functions are assumed to have the Feenberg form

$$\Psi(\mathbf{r}_1,\ldots,\mathbf{r}_N) = Ae^{[-\frac{1}{2}\sum_{i<j}u(\mathbf{r}_i-\mathbf{r}_j)-\frac{1}{2}\sum_i t(\mathbf{r}_i)]}, \tag{4}$$

including one- and two-particle correlations. The function $t(\mathbf{r})$ controls the shape of the density profile $\rho(\mathbf{r})$ associated with this wave function. It is easy to see that (4) can be rewritten in the form

$$\Psi(\mathbf{r}_1,\ldots,\mathbf{r}_N) = A\Psi_{\text{Jastrow}}(\mathbf{r}_1,\ldots,\mathbf{r}_N)\Psi_{\text{gas}}(\mathbf{r}_1,\ldots,\mathbf{r}_N), \tag{5}$$

where Ψ_{Jastrow} alone describes a bulk liquid and

$$\Psi_{\text{gas}}(\mathbf{r}_1,\ldots,\mathbf{r}_N) = \prod_{i=1}^{N}\phi_0(\mathbf{r}_i) \tag{6}$$

describes a Bose gas of N atoms all occupying the same single-particle state given by $\phi_0(\mathbf{r}) = e^{-\frac{1}{2}t(\mathbf{r})}$ (i.e., with 100% BEC). Choosing $u(\mathbf{r})$ and $t(\mathbf{r})$ appropriately, wave functions such as (4) allow one the variational freedom to describe atoms in the bulk region (where $\Psi_{\text{gas}} \simeq 1$) and in the low density surface region (note that when the atoms are far apart, one has $\Psi_{\text{Jastrow}} \simeq 1$). It is important to realize that $\phi_0(\mathbf{r})$ is not the single-particle natural orbital in (2), since the latter is defined relative to the complete wave function in (4). The interpretation of (4) as described above in (5) and (6) has not been emphasized in the previous literature on superfluid surfaces (see, however, Ref. [7]), but we think the present analysis clarifies the physics behind the complete BEC found in the surface region.

In the rest of this Letter, we discuss how our new picture can be used to describe the surface region in a rigorous fashion using the field-theoretic analysis of an inhomogeneous Bose-condensed fluid [3,5]. In addition, we point out that this has important implications concerning density functional approaches used to calculate the surface properties of superfluid ^{4}He [4,17,18].

The field-theoretical approach to Bose systems is based on the presence of the symmetry-breaking order parameter $\Phi(\mathbf{r}) \equiv \langle\hat\psi(\mathbf{r})\rangle$ as the anomalous average of the field operator $\hat\psi(\mathbf{r})$ (see Chap. 3 of Ref. [3]). The local condensate density is given by $\rho_c(\mathbf{r}) = \Phi(\mathbf{r})^2$. The equation for the order parameter can be obtained starting from the Heisenberg equation for the field operator

$$-i\frac{\partial}{\partial t}\hat\psi(\mathbf{r}) = [H', \hat\psi(\mathbf{r})], \tag{7}$$

where $H' = H - \mu N$ is the grand canonical Hamiltonian. By carrying out explicitly the commutator of (7) and taking the statistical average on the equilibrium state of the system one obtains the exact equation [6,19]

$$-\left[\frac{\hbar^2\nabla^2}{2m} - \mu\right]\Phi(\mathbf{r}) + \int d\mathbf{r}'v(\mathbf{r}-\mathbf{r}')\langle\hat\psi^\dagger(\mathbf{r}')\hat\psi(\mathbf{r}')\hat\psi(\mathbf{r})\rangle = 0, \tag{8}$$

where $v(\mathbf{r} - \mathbf{r}')$ is the bare He-He interatomic potential. When applied to the homogeneous liquid, (8) provides an exact, nontrivial relationship between the chemical potential μ and the long-range behavior of the nondiagonal two-body density matrix of a Bose-condensed system [20].

In the case of the free surface, the value of the chemical potential coincides with the bulk ^{4}He binding energy (-7.15 K at $T = 0$). In this case, (8) can be used to investigate the behavior of the order parameter $\Phi(\mathbf{r})$ when $\mathbf{r}$ is in the asymptotic, low density region far from the surface. Our reasoning is as follows. The dominant contribution to the integral in (8) comes from $\mathbf{r}'$ in the bulk region, where $\rho(\mathbf{r}')$ is large. For this contribution, $|\mathbf{r} - \mathbf{r}'|$

is large and the correlation function in (8) is given by the asymptotically exact formula

$$\begin{aligned}\langle\hat{\psi}^{\dagger}(\mathbf{r}')\hat{\psi}(\mathbf{r}')\hat{\psi}(\mathbf{r})\rangle\,|_{|\mathbf{r}-\mathbf{r}'|\to\infty} &= \langle\hat{\psi}^{\dagger}(\mathbf{r}')\hat{\psi}(\mathbf{r}')\rangle\langle\hat{\psi}(\mathbf{r})\rangle\\ &= \rho(\mathbf{r}')\Phi(\mathbf{r})\,. \end{aligned}\tag{9}$$

This may be viewed as a generalization of the Penrose-Onsager result [21]

$$\begin{aligned}\langle\hat{\psi}^{\dagger}(\mathbf{r}')\hat{\psi}(\mathbf{r})\rangle\,|_{|\mathbf{r}-\mathbf{r}'|\to\infty} &= \langle\hat{\psi}^{\dagger}(\mathbf{r}')\rangle\langle\hat{\psi}(\mathbf{r})\rangle\\ &= \Phi^{*}(\mathbf{r}')\Phi(\mathbf{r})\,. \end{aligned}\tag{10}$$

Using (9) in (8), we conclude that for $\mathbf{r}$ in the low density region (8) reduces to

$$\left[-\frac{\hbar^2\nabla^2}{2m} - \mu + \int d\mathbf{r}'v(\mathbf{r}-\mathbf{r}')\rho(\mathbf{r}')\right]\Phi(\mathbf{r}) = 0\,,\tag{11}$$

where the Hartree potential

$$v_H(\mathbf{r}) = \int d\mathbf{r}'v(\mathbf{r}-\mathbf{r}')\rho(\mathbf{r}')\tag{12}$$

describes the field at $\mathbf{r}$ due to the bulk region where the density is large. For such contributions, where $|\mathbf{r}-\mathbf{r}'|$ is large, only the long-range attractive van der Waals' tail of $v(\mathbf{r}-\mathbf{r}')$ is important. For contributions to (8) when $|\mathbf{r}-\mathbf{r}'|$ is small (i.e., when $\mathbf{r}'$ is in the low surface region), one can use $\langle\hat{\psi}^{\dagger}(\mathbf{r}')\hat{\psi}(\mathbf{r}')\hat{\psi}(\mathbf{r})\rangle \simeq \Phi(\mathbf{r}')^2\Phi(\mathbf{r})$ since we are dealing with a region of complete BEC. Such contributions are precisely those kept in the standard Gross-Pitaevskii theory [5,22] and as usual one needs to include additional multiple scattering terms [5] which screen the hard core in $v(\mathbf{r}-\mathbf{r}')$ in (8). However, the details are not important here (see Ref. [23]) since the low density at $\mathbf{r}$ makes these contributions to (8) negligible compared to the ones discussed above. Similarly, while the many-body screening of the hard core is not explicitly included in (11) and (12), contributions to $v_H(\mathbf{r})$ when $\mathbf{r}-\mathbf{r}'$ is small are negligible because of the low density at $\mathbf{r}$.

In summary, we conclude that (11) allows one to determine the exact asymptotic behavior of $\Phi(\mathbf{r})$ far from the surface of superfluid ^{4}He [23]. Interpreting (11) in physical terms, we see that the bulk of the liquid produces an effective field which acts to stabilize the dilute inhomogeneous gas in the surface region. The fact that the atoms in the low density region feel only this effective field but otherwise are not affected by their interactions explains why this region is fully Bose condensed. Equations with the same Hartree-like structure as (11) arise in discussions of the binding energy of an impurity atom on the free surface of liquid ^{4}He, based on variational wave functions [24].

For a planar surface centered at $z = 0$, the asymptotic behavior of (12) reduces at large z to $v_H(z) = -\rho_B\alpha/z^3$, where α depends only on the parameters of the He-He attractive potential [24]. Using this in (11), one finds that the order parameter has the form

$$\Phi(z) \propto e^{-(Az+B/z^2)},\tag{13}$$

where $A = \sqrt{2m|\mu|/\hbar^2} \simeq 1$ Å^{-1} and $B = m\rho_B\alpha/2\hbar^2A \simeq 5$ Å^2. The above discussion then suggests that the exponential decay [25] exhibited by the resulting density profile $\rho(z) = \rho_c(z) = |\Phi(z)|^2$ is a direct consequence of the complete BEC in the low density region, which occurs (see Fig. 1) for $\rho(z) \lesssim 0.1\rho_B$. This behavior differs, for example, from the surface density profile of a Fermi liquid, where one finds a faster decay, of the form $\rho(z) \propto z^{-2}e^{-2Az}$ [26]. Integrating (13) to find the total number of atoms in the region of essentially 100% BEC, it corresponds to an equivalent surface density of about 10^{14} atoms/cm^2.

Whatever the density is, the kinetic energy functional of a Bose-condensed system always has a contribution directly related to $\Phi(\mathbf{r})$ which is given by [5,19]

$$\int d\mathbf{r}\,\Phi^{*}(\mathbf{r})\left[-\frac{\hbar^2\nabla^2}{2m}\right]\Phi(\mathbf{r}) = \int d\mathbf{r}\,\frac{\hbar^2}{2m}|\nabla\sqrt{\rho_c(\mathbf{r})}\,|^2 + \frac{1}{2}\,m\int d\mathbf{r}\,\rho_c(\mathbf{r})\mathbf{v}_s^2(\mathbf{r})\,,\tag{14}$$

where $\Phi(\mathbf{r}) \equiv \sqrt{\rho_c(\mathbf{r})}\,e^{iS(\mathbf{r})}$ and $m\mathbf{v}_s(\mathbf{r}) \equiv \hbar\nabla S(\mathbf{r})$ is the local superfluid velocity. The expression in (14) is the origin of the kinetic energy term in (8) and (11).

The long-range coherence effects associated with superfluidity [19] are related to the local phase $S(\mathbf{r})$ of the complex (two-component) Bose order parameter $\Phi(\mathbf{r})$ defined above. In a situation like the one considered here, where the local condensate density $\rho_c(\mathbf{r})$ is rapidly decreasing normal to the surface, the superfluid flow properties may be quite different parallel and perpendicular to the surface. This clearly has experimental implications.

Phenomenological density functional theories used for inhomogeneous superfluid ^{4}He [4,17,18] always start from an energy functional of $\rho(\mathbf{r})$ alone, usually of the form

$$H[\rho(\mathbf{r})] = \int d\mathbf{r}\frac{\hbar^2}{2m}|\nabla\sqrt{\rho(\mathbf{r})}|^2 + V_{\mathrm{corr}}[\rho(\mathbf{r})]\,.\tag{15}$$

The first term is the kinetic energy of an inhomogeneous ideal Bose gas with a given local density $\rho(\mathbf{r})$, while $V_{\mathrm{corr}}[\rho(\mathbf{r})]$ is a phenomenological expression used to describe the effects of interactions. More precisely, the first term in (15) describes the energy of a fictitious liquid of N atoms as if they all occupied the same single-particle state $\phi_0(\mathbf{r}) = \sqrt{\rho(\mathbf{r})/N}$, corresponding to $\Phi(\mathbf{r}) = \sqrt{\rho(\mathbf{r})}$. A complete density functional theory of Bose-condensed liquids would involve functionals of both the local density $\rho(\mathbf{r})$ and the local order parameter $\Phi(\mathbf{r})$ [27]. In such a theory, the first term of (14) would naturally arise in place of the first term in (15), although we note that the two terms coincide in the important low density surface region since $\rho_c(\mathbf{r}) \simeq \rho(\mathbf{r})$.

It would be useful to have finite temperature path integral Monte Carlo calculations of $\rho_c(\mathbf{r})$ in the surface region of superfluid ^{4}He [28]. Physically, it seems clear that the surface region will Bose condense at the bulk superfluid transition temperature $T_\lambda \simeq 2.17$ K. The chemical potential is the same for the liquid and vapor in equilibrium and since it is large and negative ($\mu \simeq -7$ K from 0 to 2 K), the ^{4}He vapor can always be described as an ideal classical gas. Figure 1 is valid only at low temperatures ($\lesssim 1$ K). As the temperature increases, the condensate density $\rho_c(\mathbf{r})$ will be increasingly depleted [3] and thus it will be a smaller component of the total local density $\rho(\mathbf{r})$, in *both* the bulk and surface regions. However, we expect that $\rho_c(\mathbf{r})$ will still decrease exponentially in the surface region, while $\rho(\mathbf{r})$ will go over smoothly to the small but finite density of the ^{4}He vapor.

In conclusion, in this Letter we have argued that the surface region of superfluid ^{4}He in self-bound phases can be described as an inhomogeneous dilute Bose gas which is completely Bose condensed at $T = 0$. Dramatic evidence for this "self-bound Bose gas" is given by the numerical calculations in Ref. [7]. We have noted that the asymptotic low density surface region can be described *exactly* by the order parameter $\Phi(\mathbf{r})$ given by the solution of a generalized Gross-Pitaevskii equation (11) taking full account of the long-range van der Waals tail of the He-He potential. Optical excitation of the surface atoms might be a way of detecting the completely Bose-condensed nature of the surface region of superfluid ^{4}He. We have used our new physical ideas to suggest a more microscopic basis for the kinetic energy term used in density functional theories. This inhomogeneous Bose-condensed gas is readily available, and should complement the more exotic Bose gas systems on which much current research has concentrated [29].

We have had useful discussions concerning these results with F. Laloë and P. Nozières. A. G. would like to thank the Università di Trento for financial support and stimulating hospitality during a sabbatical leave. His research was also supported by NSERC of Canada.

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