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Bose Condensation and Momentum Distribution in a Simplified Liquid-^{4}He Ground State*

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Bose-Einstein condensation and the atomic momentum distribution are investigated for a simplified liquid ^{4}He ground state obtained by antisymmetrization, with respect to the electron variables, of a product of ^{4}He-atom wave functions, each in its individual electronic and translational ground state. The momentum distribution is defined and evaluated by the method of redundant modes and generalized Tani transformation. Bose-Einstein condensation is found, with a condensate depletion of 6% owing to the purely kinematical repulsion arising from the Pauli exclusion principle. The noncondensed atoms are spread in momentum space by an amount of a few inverse Bohr radii. Dynamical correlations would increase the depletion in a more realistic ground state.

I. INTRODUCTION

It was originally suggested by London,[1] and is now almost universally accepted, that the λ transition of liquid ^{4}He, and its superfluidity and other exotic properties below the λ-transition temperature, are associated with Bose-Einstein condensation of the helium atoms in momentum space. It is also known that such Bose-Einstein condensation is in a certain sense equivalent to "off-diagonal long-range order" (ODLRO) of the appropriate reduced density matrix.[2-5] It is customary in microscopic theories of liquid ^{4}He to regard ^{4}He atoms as little Bose billiard balls. It is well known that the mutual impenetrability of pairs of atoms arises from the effects of the exclusion principle together with the filled-shell electronic configurations, and that the long-range attraction (van der Waals force) which makes the system a liquid arises from coupled virtual electromagnetic transitions of pairs of atoms. However, in conventional treatments such effects are simulated by a suitable phenomenological potential acting between pairs of "elementary" bosons, without introducing electronic degrees of freedom into the model explicitly. The λ transition is then viewed as a Bose-Einstein condensation in a system of interacting but "elementary" bosons. In such a model, the definition of atomic momentum distribution and Bose-Einstein condensation is straightforward and essentially unique, and the associated ODLRO occurs in the single-boson reduced density matrix.[2-5]

In some cases it is, however, advantageous to introduce the electronic degrees of freedom into the description of liquid ^{4}He. This is, in fact, essential in a first-principles approach to the theory of excitons, and more generally in theories of various types of excitations of the liquid which arise from or are related to electronic excitations of its individual atoms. Furthermore, inclusion of the electronic degrees of freedom offers some advantages even in the description of the ground state, since use of properly antisymmetrized wave functions then incorporates the short-range interatomic repulsion purely kinematically, without the necessity of employing phenomenological hard-core potentials, which are difficult to deal with mathematically.

As soon as the electronic degrees of freedom are introduced explicitly, one is faced with the problem of defining the atomic momentum distribution; the standard definitions of momentum distribution and Bose-Einstein condensation, e.g., in terms of second quantization, apply only to *elementary* bosons. Any definition of occupation numbers of states of *composite* particles (e.g., atoms) necessarily involves a generalization, and is therefore nonunique. Several definitions, not all equivalent, have been proposed.[6-9] The one which seems to us to be conceptually simplest, and also most closely related to the standard definition for elementary particles, is based on the familiar idea of the introduction of initially redundant variables corresponding to the composite particles, followed by a canonical transformation (here a "generalized Tani transformation"[9]) which gives these extra modes physical content. Such an approach, which will be employed here, is similar to that of the Bohm-Pines theory of plasma oscillations.[10] We shall apply this method to the evaluation of the atomic momentum distribution of a simplified liquid-^{4}He ground state obtained by antisymmetrization, with respect to the electron variables, of a product of ^{4}He-atom wave functions, each in its individual electronic and translational ground state. Some of the properties of such a state were investigated previously.[11] The atomic pair-distribution function was found to vanish at zero interatomic separation, as a purely kinematical result of the exclusion principle together with the closed-shell structure of the helium atoms. Also, the single ^{4}He-atom reduced-density matrix (including electronic variables) was found to exhibit ODLRO of a type shown by Kohn and Sherrington[5] to imply nonclassical rotational behavior expected in a superfluid. The momentum distribution was also investigated, but using a different definition[6] of atomic occupation numbers. The purpose of the present paper is to reinvestigate the momentum distribution and Bose-Einstein condensation of the same many-atom state, using the definition[9] of atomic occupation numbers which we now believe to be most appropriate.

II. FORMULATION

The representation which we shall employ has been described previously[9] for the slightly simpler case of hydrogen atoms. In terms of the standard quantized field operators $\psi(\vec{R})$ for ^{4}He nuclei (α particles) and $\psi(x)$ for electrons, one can write the state vector of a single ^{4}He atom with wave function $\varphi_\alpha(\vec{R}x_1x_2)$ in the form

$$|\varphi_\alpha\rangle = A_\alpha{}^\dagger|0\rangle \tag{1}$$

where $|0\rangle$ is the normalized vacuum state and[9,11]

$$A_\alpha{}^\dagger = 2^{-1/2}\int d^3R\, dx_1 dx_2\, \varphi_\alpha(\vec{R}x_1x_2) \times \psi^\dagger(\vec{R})\psi^\dagger(x_1)\psi^\dagger(x_2). \tag{2}$$

Using the elementary Bose commutation relations of the nucleus operators, the Fermi anticommutation relations of the electron operators, and assuming the atomic states φ_α orthonormal, one can show that

$$\begin{aligned} &[A_\alpha, A_\beta] = 0, \quad [A_\alpha, A_\beta{}^\dagger] = \delta_{\alpha\beta} + C_{\alpha\beta}, \\ &[\psi(\vec{R}), A_\alpha] = [\psi(x), A_\alpha] = 0, \\ &[\psi(\vec{R}), A_\alpha{}^\dagger] = 2^{-1/2}\int dx_1 dx_2\, \varphi_\alpha(\vec{R}x_1x_2)\psi^\dagger(x_1)\psi^\dagger(x_2), \\ &[\psi(x), A_\alpha{}^\dagger] = 2^{1/2}\int d^3R\, dx'\, \varphi_\alpha(\vec{R}xx')\psi^\dagger(\vec{R})\psi^\dagger(x'). \end{aligned} \tag{3}$$

Here $C_{\alpha\beta}$ is a sum of terms of the structures $\psi^\dagger\psi$ and $\psi^\dagger\psi^\dagger\psi\psi$, and exhibits the kinematical effect of the composite structure of helium atoms. Its explicit form will not be required here, but is is important to realize that the A_α and $A_\alpha{}^\dagger$ operators do not satisfy elementary Bose commutation relations owing to the terms $C_{\alpha\beta}$. Similarly, the nonvanishing commutators $[\psi(\vec{R}), A_\alpha{}^\dagger]$ and $[\psi(x), A_\alpha{}^\dagger]$ exhibit the lack of kinematical independence of ^{4}He atoms from α particles and electrons. A state of many ^{4}He atoms can be represented[12] as a sum of atomic product states $A_{\alpha_1}{}^\dagger \cdots A_{\alpha_n}{}^\dagger|0\rangle$ but such a representation is difficult to work with because of the complicated commutation relations (3).

Now introduce the ideal atom space, a Hilbert space independent of the physical state space. It consists of normalizable linear combinations of ideal atomic product states $a_{\alpha_1}{}^\dagger \cdots a_{\alpha_n}{}^\dagger|0\rangle$ where, by definition, the a_α and $a_\alpha{}^\dagger$ satisfy elementary Bose commutation relations and are kinematically independent of the nucleus and electron fields:

$$\begin{aligned} &[a_\alpha, a_\beta] = 0, \quad [a_\alpha, a_\beta{}^\dagger] = \delta_{\alpha\beta}, \\ &[\psi(\vec{R}), a_\alpha] = [\psi(x), a_\alpha] = [\psi(\vec{R}), a_\alpha{}^\dagger] = [\psi(x), a_\alpha{}^\dagger] = 0. \end{aligned} \tag{4}$$

If we define the ideal state space to be the direct product of the physical and ideal-atom state spaces, then the physical state space is isomorphic with the space of those ideal states $|\psi\rangle$ satisfying the constraints

$$a_\alpha|\psi\rangle = 0, \quad \text{all } \alpha. \tag{5}$$

In other words, there are no ideal atoms in states $|\psi\rangle$, so that the ideal atoms are "redundant modes."

Physical content can be given to these redundant modes by use of an appropriate unitary transformation, the generalized Tani transformation

$$U = e^{(\pi/2)F}, \quad F = \sum_\alpha (a_\alpha{}^\dagger A_\alpha - A_\alpha{}^\dagger a_\alpha). \tag{6}$$

It can be shown[9] that

$$UA_\alpha{}^\dagger|0\rangle = a_\alpha{}^\dagger|0\rangle; \tag{7}$$

i.e., a state of one physical atom transforms exactly into a state of one ideal atom. Such simple transformation properties cannot be expected for states of more than one atom, since a system of real atoms cannot be unitarily equivalent to a system of "elementary" ones. In the approximation in which all atomic overlap is neglected, and hence $C_{\alpha\beta}$ and the commutators $[\psi(\vec{R}), A_\alpha{}^\dagger]$ and $[\psi(x), A_\alpha{}^\dagger]$ in (3) are dropped, one has

$$UA_{\alpha_1}{}^\dagger \cdots A_{\alpha_n}{}^\dagger|0\rangle \approx a_{\alpha_1}{}^\dagger \cdots a_{\alpha_n}{}^\dagger|0\rangle. \tag{8}$$

However, the corrections to (8) are not negligible in liquid ^{4}He, in view of the importance of exchange and hence the non-negligibility of atomic overlap in this system. Nevertheless, the transformation (6) is still useful, since it can be used to introduce the easily interpreted and easily manipulated ideal-atom operators a_α and $a_\alpha{}^\dagger$. If A is any observable and $|\psi\rangle$ and $|\psi'\rangle$ any states in the ideal state space satisfying (5), then

$$\langle\psi|A|\psi'\rangle = (\psi|U^{-1}AU|\psi), \tag{9}$$

where, for any $|\psi\rangle$, the transformed state $|\psi)$ is defined as[9]

$$|\psi) = U^{-1}|\psi\rangle. \tag{10}$$

In the transformed state $|\psi)$ and transformed observable $U^{-1}AU$, atoms are represented by true Bose operators a_α and $a_\alpha{}^\dagger$. Thus, it is natural to define the atomic occupation numbers n_α by

$$n_\alpha = (\psi| a_\alpha{}^\dagger a_\alpha |\psi) = \langle \psi | U a_\alpha{}^\dagger a_\alpha U^{-1} |\psi\rangle . \tag{11}$$

In contrast with the operators $A_\alpha{}^\dagger A_\alpha$, the operators $a_\alpha{}^\dagger a_\alpha$ are true Bose occupation-number operators and have integral eigenvalues, so that the definition (11) is quite natural. There are two exact sum rules which lend further credence to this definition. Define the total atom number operator $N_{\rm at}$, total nucleus number operator $N_{\rm nuc}$, and total electron-number operator $N_{\rm elec}$ in the standard way:

$$\begin{aligned} N_{\rm at} &= \sum_\alpha a_\alpha{}^\dagger a_\alpha , \\ N_{\rm nuc} &= \int d^3R\, \psi^\dagger(\vec{\rm R})\psi(\vec{\rm R}) , \\ N_{\rm elec} &= \int dx\, \psi^\dagger(x)\psi(x). \end{aligned} \tag{12}$$

Then it is easy to see that[9]

$$[(N_{\rm at} + N_{\rm nuc}), U] = 0, \quad [(2N_{\rm at} + N_{\rm elec}), U] = 0, \tag{13}$$

since F creates an ideal atom whenever it destroys a nucleus, and creates an ideal atom whenever it destroys two electrons, and vice versa. If $|\psi\rangle$ is any state of n nuclei and $2n$ electrons satisfying (5), e.g., any state of n physical ^{4}He atoms, then

$$\begin{aligned} (N_{\rm at} + N_{\rm nuc})|\psi\rangle &= n|\psi\rangle , \\ (2N_{\rm at} + N_{\rm elec})|\psi\rangle &= 2n|\psi\rangle . \end{aligned} \tag{14}$$

As far as the states $|\psi\rangle$ are concerned, these relations are trivial. However, they acquire nontrivial and useful content in terms of the transformed states $|\psi)$. By (10) and (13), one has also

$$\begin{aligned} (N_{\rm at} + N_{\rm nuc})|\psi) &= n|\psi) , \\ (2N_{\rm at} + N_{\rm elec})|\psi) &= 2n|\psi). \end{aligned} \tag{15}$$

The physical significance is that $|\psi)$ the total number of nuclei is the sum of the number of bound atoms (with occupation numbers $a_\alpha{}^\dagger a_\alpha$) and the number of free nuclei [with number-density operator $\psi^\dagger(\vec{\rm R})\psi(\vec{\rm R})$]; similarly, the total number of electrons is *twice* the number of bound atoms plus the number of free electrons. This supports the interpretation of the definition (11) of the atomic occupation numbers n_α, and suggests similar definitions of the numbers $n_{\rm nuc}$ and $n_{\rm elec}$ of free nuclei and electrons:

$$\begin{aligned} n_{\rm nuc} &= (\psi|N_{\rm nuc}|\psi) = \langle\psi|U N_{\rm nuc} U^{-1}|\psi\rangle , \\ n_{\rm elec} &= (\psi|N_{\rm elec}|\psi) = \langle\psi|U N_{\rm elec} U^{-1}|\psi\rangle . \end{aligned} \tag{16}$$

In view of (15), one has the sum rules

$$\sum_\alpha n_\alpha + n_{\rm nuc} = n, \quad 2\sum_\alpha n_\alpha + n_{\rm elec} = 2n. \tag{17}$$

For liquid ^{4}He in its ground state or more generally in equilibrium, ionized states have very small occupations, so that (17) expresses the fact that the numbers of atoms in various single-atom states φ_α must sum to n.

The transforms needed to evaluate (11) can be found by the equation-of-motion method. Define, for any operator A,

$$A(\epsilon) = e^{-\epsilon F} A e^{\epsilon F}, \tag{18}$$

so that by (6)

$$A(0) = A, \quad A(-\tfrac{1}{2}\pi) = UAU^{-1}. \tag{19}$$

Then $A(\epsilon)$ satisfies the "equation of motion"

$$\frac{dA(\epsilon)}{d\epsilon} = e^{-\epsilon F}[A, F]\, e^{\epsilon F} = [A(\epsilon), F(\epsilon)]. \tag{20}$$

In particular, it follows from (3), (4), and (6) that

$$\begin{aligned} \frac{da_\alpha(\epsilon)}{d\epsilon} &= A_\alpha(\epsilon), \\ \frac{dA_\alpha(\epsilon)}{d\epsilon} &= -a_\alpha(\epsilon) - \sum_\beta C_{\alpha\beta}(\epsilon)\, a_\beta(\epsilon), \\ \frac{\partial\psi(\vec{\rm R},\epsilon)}{\partial\epsilon} &= -2^{-1/2}\sum_\alpha \int dx_1\, dx_2\, \varphi_\alpha(\vec{\rm R}\, x_1 x_2) \\ &\qquad \times \psi^\dagger(x_1,\epsilon)\psi^\dagger(x_2,\epsilon)\, a_\alpha(\epsilon), \\ \frac{\partial\psi(x,\epsilon)}{\partial\epsilon} &= -2^{1/2}\sum_\alpha \int d^3R\, dx'\, \varphi_\alpha(\vec{\rm R}, xx') \\ &\qquad \times \psi^\dagger(\vec{\rm R},\epsilon)\psi^\dagger(x',\epsilon)\, a_\alpha(\epsilon). \end{aligned} \tag{21}$$

These equations and their Hermitian conjugates are a set of coupled nonlinear integrodifferential equations, and cannot be solved in closed form. However, the lowest-order solutions, valid when the density of the system is sufficiently low (as it is for liquid ^{4}He), can be found by iteration. In the approximation in which the atoms behave like elementary bosons, and hence the terms proportional to $C_{\alpha\beta}(\epsilon)$ in the differential equation for $A_\alpha(\epsilon)$ and the entire right-hand sides of the differential equations for $\psi(\vec{\rm R},\epsilon)$ and $\psi(x,\epsilon)$ are dropped, one has

$$\begin{aligned} &\frac{da_\alpha(\epsilon)}{d\epsilon} = A_\alpha(\epsilon), \quad \frac{dA_\alpha(\epsilon)}{d\epsilon} \approx -a_\alpha(\epsilon), \\ &\frac{\partial\psi(\vec{\rm R},\epsilon)}{\partial\epsilon} \approx 0, \quad \frac{\partial\psi(x,\epsilon)}{\partial\epsilon} \approx 0. \end{aligned} \tag{22}$$

The solutions satisfying the correct "initial" ($\epsilon = 0$) conditions (19) are

$$\begin{aligned} a_\alpha(\epsilon) &\approx a_\alpha \cos\epsilon + A_\alpha \sin\epsilon, \\ A_\alpha(\epsilon) &\approx A_\alpha \cos\epsilon - a_\alpha \sin\epsilon, \\ \psi(\vec{\rm R},\epsilon) &\approx \psi(\vec{\rm R}), \\ \psi(x,\epsilon) &\approx \psi(x). \end{aligned} \tag{23}$$

Putting $\epsilon = -\frac{1}{2}\pi$ and noting that $U^{-1}|0\rangle = |0\rangle$, one easily verifies (8), since by (19) one has $UA_\alpha U^{-1} = A_\alpha(-\frac{1}{2}\pi)$. More accurate solutions can be obtained by substitution of these lowest-order approximations into the right-hand sides of (21) and solution of the resultant inhomogeneous linear equations. Putting $\epsilon = -\frac{1}{2}\pi$ in the resultant expressions, one finds

$$U a_\alpha U^{-1} = -A_\alpha - \sum_\beta C_{\alpha\beta}(\tfrac{1}{4}\pi a_\beta - \tfrac{1}{2}A_\beta) + \cdots ,$$

$$U\psi(\vec{\mathrm{R}})U^{-1} = \psi(\vec{\mathrm{R}}) + 2^{-1/2}\sum_\alpha \int dx_1\, dx_2\, \varphi_\alpha(\vec{\mathrm{R}}x_1 x_2) \times \psi^\dagger(x_1)\psi^\dagger(x_2)(a_\alpha - A_\alpha) + \cdots ,$$

$$U\psi(x)U^{-1} = \psi(x) + 2^{1/2}\sum_\alpha \int d^3R\, dx'\, \varphi_\alpha(\vec{\mathrm{R}}xx') \times \psi^\dagger(\vec{\mathrm{R}})\psi^\dagger(x')(a_\alpha - A_\alpha) + \cdots . \tag{24}$$

It is now possible to write the expressions (11) and (16) in more explicit form. Making use of (24), (2), (5), and (12), one finds

$$\begin{aligned} n_\alpha &= \langle\psi|A_\alpha{}^\dagger A_\alpha|\psi\rangle - \tfrac{1}{2}\sum_\beta (\langle\psi|A_\alpha{}^\dagger C_{\alpha\beta}A_\beta|\psi\rangle + \mathrm{c.c.}) \\ &\quad + \tfrac{1}{4}\sum_{\beta\gamma}\langle\psi|A_\beta{}^\dagger C_{\alpha\beta}{}^\dagger C_{\alpha\gamma}A_\gamma|\psi\rangle + \cdots , \\ n_{\mathrm{nuc}} &= \langle\psi|N_{\mathrm{nuc}}|\psi\rangle - \sum_\alpha \langle\psi|A_\alpha{}^\dagger A_\alpha|\psi\rangle + \cdots , \\ n_{\mathrm{elec}} &= \langle\psi|N_{\mathrm{elec}}|\psi\rangle - 2\sum_\alpha \langle\psi|A_\alpha{}^\dagger A_\alpha|\psi\rangle + \cdots . \end{aligned} \tag{25}$$

Since $|\psi\rangle$ is a state of n ^{4}He atoms, it satisfies

$$\langle\psi|N_{\mathrm{nuc}}|\psi\rangle = n, \quad \langle\psi|N_{\mathrm{elec}}|\psi\rangle = 2n. \tag{26}$$

Hence the expressions (25) are consistent, within the order to which they are evaluated, with the exact sum rules (17). In view of these sum rules, it is not necessary to evaluate n_{nuc} and n_{elec} separately; one needs only the atomic occupation numbers n_α. It is convenient to express them in terms of the reduced density matrices of the state $|\psi\rangle$. The single-atom (one nucleus, two electron) density matrix ρ_3 is defined by

$$\rho_3(\vec{\mathrm{R}}x_1 x_2,\ \vec{\mathrm{R}}'x_1' x_2') = \langle\psi|\psi^\dagger(\vec{\mathrm{R}}')\psi^\dagger(x_1')\psi^\dagger(x_2')\psi(x_2)\psi(x_1)\psi(\vec{\mathrm{R}})|\psi\rangle . \tag{27}$$

The first term in the expression (25) for n_α is proportional to ρ_3. Recalling that $C_{\alpha\beta}$ is a sum of terms of the structures $\psi^\dagger\psi$ and $\psi^\dagger\psi^\dagger\psi\psi$, one sees that the remaining terms in n_α are proportional to $\rho_4, \rho_5, \ldots$; such terms are expected to be small compared to those involving ρ_3 at the low density of liquid ^{4}He, and will not be evaluated here. One thus has with (2)

$$\begin{aligned} n_\alpha &= \tfrac{1}{2}(\varphi_\alpha, \rho_3\varphi_\alpha) + \cdots \\ &\equiv \tfrac{1}{2}\int \varphi_\alpha^*(\vec{\mathrm{R}}x_1 x_2)\rho_3(\vec{\mathrm{R}}x_1 x_2, \vec{\mathrm{R}}'x_1' x_2') \\ &\quad \times \varphi_\alpha(\vec{\mathrm{R}}'x_1' x_2')\, d^3R\, dx_1\, dx_2\, d^3R'\, dx_1'\, dx_2' + \cdots . \end{aligned} \tag{28}$$

It is amusing to note that within the approximation of including only terms in ρ_3, expression (28) differs from the previous expression[13] (based on a different definition of atomic occupation numbers) only in normalization. The difference is, nevertheless, crucial—the previous expression being smaller by the factor $[n(2n-1)]^{-1} \sim 10^{-46}$! The reason for this enormous difference in normalization is that in the previous definition[6] both bound and continuum (unbound) states were treated as atomic states. On the other hand, according to the new definition (11) upon which (28) is based, only *bound* states of one nucleus and two electrons are regarded as atomic states φ_α, so that the set $\{\varphi_\alpha\}$ is undercomplete. The sum rules (17) are then completed by the contributions n_{nuc} and n_{elec} of *free* (unbound) nuclei and electrons. We regard this approach as more physical than the old one. This will be confirmed by the subsequent evaluation of (28) for a simplified liquid-^{4}He ground state, which will be found to lead to Bose-Einstein condensation in the usual sense (n_0 of order n). It will also be found that n_{nuc} and n_{elec} are negligibly small, in agreement with the physical expectation that ionized states have very small occupation in liquid ^{4}He in its ground state.

III. SIMPLIFIED LIQUID-^{4}He GROUND STATE

In this section we shall briefly review the definition and properties of the simplified liquid-^{4}He ground state[11] $|\psi_0\rangle$ for which (28) will be evaluated.

Let $A_0{}^\dagger$ be the creation operator for a single ^{4}He atom in its ground state (zero total linear momentum and electronic ground state), obtained by taking φ_0 in (2) to be the corresponding ^{4}He-atom ground-state wave function. The closest analog of the ideal Bose-gas ground state which can be constructed from n real ^{4}He atoms can then be written in the form

$$\mathrm{const} \times (A_0{}^\dagger)^n|0\rangle . \tag{29}$$

This is the second-quantized form of the Schrödinger wave function

$$\mathrm{const} \times A_{2n}[\varphi_0(\vec{\mathrm{R}}_1 x_1 x_2)\varphi_0(\vec{\mathrm{R}}_2 x_3 x_4)\cdots \times \varphi_0(\vec{\mathrm{R}}_n x_{2n-1} x_{2n})] , \tag{30}$$

where A_{2n} is the antisymmetrizer with respect to the $2n$ electronic variables; this state is then automatically symmetrical under permutations of the nuclear variables $\vec{\mathrm{R}}_1 \cdots \vec{\mathrm{R}}_n$, so that a nuclear symmetrizer is not necessary. Although the state (29) does not contain dynamical interatomic correlations, it does incorporate kinematical correlations implied by the exclusion principle, which acts to inhibit overlap of filled atomic shells.[11] The state (29) is somewhat analogous to the $2n$-electron projection of the ground state of the BCS theory of superconductivity,[14] which is also of the form (29) with $A_0{}^\dagger$ the creation operator for a Cooper pair.

As in the BCS theory, it is mathematically convenient to slightly relax the conservation of total particle number. Thus we replace (29) by the state

$$|\psi_0\rangle = S|0\rangle, \quad S = e^G, \qquad G = c(A_0{}^\dagger - A_0). \tag{31}$$

This state is easier to work with since it is expressed in terms of a unitary transformation S. The real positive constant c is to be determined so as to give the correct total number n of ^{4}He atoms (hence n nuclei) in the sense of an expectation value:

$$\langle\psi_0|N_{\text{nuc}}|\psi_0\rangle = n. \tag{32}$$

In analogy with the BCS theory, the fractional fluctuations in N_{nuc} (hence in the number of atoms) can be shown[11] to be of order $n^{-1/2}$ for $n\to\infty$, and hence negligible. The approximation involved in the replacement of (29) by (31) is therefore quite innocuous. The calculations are further simplified by adopting a simple Hartree-Fock form for the atomic ground state φ_0, which implies that $A_0{}^\dagger$ can be written in the form[11]

$$A_0{}^\dagger = \Omega^{-1/2} \int d^3R\, c_\uparrow{}^\dagger(\vec{R})\, c_\downarrow{}^\dagger(\vec{R})\, \psi^\dagger(\vec{R}), \tag{33}$$

where Ω is the volume of the system,[15] $\psi^\dagger(\vec{R})$ creates a nucleus at $\vec{R}$, and $c_\sigma{}^\dagger(\vec{R})$ ($\sigma = \uparrow$ or $\downarrow$) creates an electron of spin z-component σ in the Hartree-Fock orbital u_0 centered on the nucleus at $\vec{R}$,

$$c_\sigma{}^\dagger(\vec{R}) = \int d^3r\, u_0(\vec{r} - \vec{R})\, \psi_\sigma{}^\dagger(\vec{r}). \tag{34}$$

Here $\psi_\sigma{}^\dagger(\vec{r})$ is the creation operator for an electron of spin σ at position $\vec{r}$. It follows from the usual anticommutation relations for the ψ_σ and $\psi_\sigma{}^\dagger$ that the operators (34) satisfy anticommutation relations

$$\{c_\sigma(\vec{R}), c_{\sigma'}(\vec{R}')\} = 0, \qquad \{c_\sigma(\vec{R}), c_{\sigma'}{}^\dagger(\vec{R}')\} = \delta_{\sigma\sigma'}\, w(\vec{R} - \vec{R}'), \tag{35}$$

where w is the interatomic overlap integral

$$w(\vec{R}) = \int u_0{}^*(\vec{r})\, u_0(\vec{r} + \vec{R})\, d^3r. \tag{36}$$

Both u_0 and w are spherically symmetric[16] and of range $\sim a_0$, where a_0 is the Bohr radius.

It has already been shown elsewhere[11] that the state (31) has the following properties: (i) the single-atom density matrix ρ_3 [Eq. (27)] exhibits ODLRO; i.e., it contains a non-negligible separable term $\chi(\vec{R}x_1x_2)\chi^*(\vec{R}'x_1'x_2')$ and hence does not vanish at infinite atomic separation; (ii) the nuclear pair distribution function $D(R_{12})$, the probability of finding two nuclei (hence two atoms) with separation R_{12}, vanishes as $R_{12}\to 0$; (iii) the *nuclear* momentum distribution is a smooth function of range $\sim\hbar/a_0$ in momentum space, and does not exhibit Bose-Einstein condensation. Property (i) implies, by an argument of Kohn and Sherrington,[5] that the system will exhibit nonclassical behavior under rotation, as expected for a superfluid. Property (ii) demonstrates that part of the "hard-core" interatomic repulsion is kinematically built into the state (31) as a result of the Pauli exclusion principle which inhibits overlap of filled electronic shells. Property (iii) is expected because of the motion of an atomic nucleus in response to that of the electrons of the same atom (conservation of momentum); as a result, any Bose-Einstein condensation of the atoms is "smeared" and not exhibited in the momentum distribution of the nuclei. In fact, we shall find in Sec. IV that the *atomic* momentum distribution, defined in terms of (28), does exhibit Bose-Einstein condensation.

IV. ATOMIC MOMENTUM DISTRIBUTION AND BOSE CONDENSATION

By comparison of (2) with (27) and (28) one sees that the atomic occupation-number distribution of the state $|\psi_0\rangle$ can be expressed in the form

$$n_\alpha = \langle\psi_0|A_\alpha{}^\dagger A_\alpha|\psi_0\rangle + \cdots. \tag{37}$$

The index α of the atomic states φ_α can be decomposed as $\alpha = (\vec{k}, \nu)$ where $\vec{k}$ is the translational wave vector, related to the total linear momentum $\vec{p}$ of the atom by $\vec{p} = \hbar\vec{k}$, and ν is the set of all internal quantum numbers labeling the electronic state. For the time being we shall consider only states which are not internally excited, and hence differ from φ_0 only by having a momentum which is in general nonzero. Denote the wave functions and creation operators for such states by $\varphi_{\vec{k}}$ and $A_{\vec{k}}{}^\dagger$, and the corresponding occupation number by $n_{\vec{k}}$, which then has the interpretation of the momentum distribution of atoms in their internal ground states. The $\varphi_{\vec{k}}$ differ from φ_0 only by a phase factor[17] $e^{i\vec{k}\cdot\vec{R}}$ corresponding to the momentum $\hbar\vec{k}$. Accordingly, (33) generalizes to

$$A_{\vec{k}}{}^\dagger = \Omega^{-1/2} \int d^3R\, e^{i\vec{k}\cdot\vec{R}}\, c_\uparrow{}^\dagger(\vec{R})\, c_\downarrow{}^\dagger(\vec{R})\, \psi^\dagger(\vec{R}). \tag{38}$$

Insertion into (37) then yields

$$n_{\vec{k}} = \Omega^{-1} \int e^{i\vec{k}\cdot(\vec{R}'-\vec{R})} \times \rho_3(\vec{R}\uparrow, \vec{R}\downarrow, \vec{R}; \vec{R}'\uparrow, \vec{R}'\downarrow, \vec{R}')\, d^3R\, d^3R' + \cdots, \tag{39}$$

where the ρ_3 in (39), not to be confused with that of (27) (although it is closely related), is

$$\rho_3(\vec{R}\uparrow,\vec{R}\downarrow,\vec{R};\ \vec{R}'\uparrow,\vec{R}'\downarrow,\vec{R}')=\langle\psi_0|c_\uparrow{}^\dagger(\vec{R}')c_\downarrow{}^\dagger(\vec{R}')\psi^\dagger(\vec{R}')\psi(\vec{R})c_\downarrow(\vec{R})c_\uparrow(\vec{R})|\psi_0\rangle$$
$$=\langle 0|\,[c_\uparrow'(\vec{R}')]^\dagger[c_\downarrow'(\vec{R}')]^\dagger[\psi'(\vec{R}')]^\dagger\,\psi'(\vec{R})c_\downarrow'(\vec{R})c_\uparrow'(\vec{R})|0\rangle, \tag{40}$$

with

$$c_\sigma'(\vec{R})=S^{-1}c_\sigma(\vec{R})S,\quad \psi'(\vec{R})=S^{-1}\psi(\vec{R})S. \tag{41}$$

Since the state $|\psi_0\rangle$ is an eigenstate of total linear momentum with eigenvalue zero, ρ_3 is translationally invariant, i.e.,

$$\rho_3(\vec{R}\uparrow,\vec{R}\downarrow,\vec{R};\ \vec{R}'\uparrow,\vec{R}'\downarrow,\vec{R}')$$
$$=\rho_3(\vec{R}-\vec{R}',\uparrow,\vec{R}-\vec{R}',\downarrow,\vec{R}-\vec{R}';\ 0\uparrow,0\downarrow,0). \tag{42}$$

Hence, one of the integrations in (39) can be performed immediately:

$$n_{\vec{k}}=\int\rho_3(\vec{R}\uparrow,\vec{R}\downarrow,\vec{R};\ 0\uparrow,0\downarrow,0)\,e^{-i\vec{k}\cdot\vec{R}}\,d^3R. \tag{43}$$

By insertion of (34) into (40), one can express ρ_3 in terms of the density matrix D_3 previously evaluated[18]:

$$\rho_3(\vec{R}\uparrow,\vec{R}\downarrow,\vec{R};\ 0\uparrow,0\downarrow,0)=|\chi|^2+\rho_3'(\vec{R}\uparrow,\vec{R}\downarrow,\vec{R};\ 0\uparrow,0\downarrow,0),$$
$$\rho_3'(\vec{R}\uparrow,\vec{R}\downarrow,\vec{R};\ 0\uparrow,0\downarrow,0)=\rho^2[\rho_3{}^{(1)}(\vec{R})+\rho_3{}^{(2)}(\vec{R})]+\cdots,$$
$$\rho_3{}^{(1)}(\vec{R})=-8w(\vec{R})\,a_0{}^3I_{13}(\vec{R})+w^2(\vec{R})\,a_0{}^3I_{22}(\vec{R}),$$
$$\rho_3{}^{(2)}(\vec{R})=2w(\vec{R})\,a_0{}^3I_{11}(\vec{R})+2a_0{}^3I_{22}(\vec{R}). \tag{44}$$

Here the integrals $I_{\mu\nu}$ are defined by

$$a_0{}^3I_{\mu\nu}(\vec{R})=\int[w(\vec{R}-\vec{R}')]^\mu[w(\vec{R}')]^\nu\,d^3R', \tag{45}$$

and the terms "$\cdots$" not exhibited are of higher order in the small parameter $\rho a_0{}^3$, where $\rho=n/\Omega$ is the number density of ^{4}He atoms and a_0 is the Bohr radius. For liquid ^{4}He under normal experimental conditions, $\rho a_0{}^3=0.0033$. The quantity χ in (44), the analog of the "pairing amplitude" of BCS theory, is, by translational invariance of $|\psi_0\rangle$, independent of $\vec{R}$ and closely related to the condensate wave function $\chi(x_1x_2\vec{R})$ previously defined,[19] associated with ODLRO of D_3, hence of ρ_3. It can be expressed in terms of integrals x and y previously defined[20]:

$$\chi=\rho^{1/2}[1-(x-\tfrac{1}{2}y)\rho a_0{}^3+O((\rho a_0{}^3)^2)], \tag{46}$$

with

$$xa_0{}^3=\int w^2(\vec{R})\,d^3R,\quad ya_0{}^3=\int w^4(\vec{R})\,d^3R. \tag{47}$$

In accordance with the decomposition of ρ_3 into the ODLRO term $|\chi|^2$ and the residue ρ_3', $n_{\vec{k}}$ [Eq. (42)] decomposes into a Bose condensate contribution n_0 and an uncondensed contribution $n_{\vec{k}}'$:

$$n_{\vec{k}}=n_0\,\delta_{\vec{k}0}+n_{\vec{k}}', \tag{48}$$

with

$$n_0=|\chi|^2\Omega=n[1-(2x-y)\rho a_0{}^3+O((\rho a_0{}^3)^2)] \tag{49}$$

and

$$n_{\vec{k}}'=\int\rho_3'(\vec{R}\uparrow,\vec{R}\downarrow,\vec{R};\ 0\uparrow,0\downarrow,0)\,e^{-i\vec{k}\cdot\vec{R}}\,d^3R. \tag{50}$$

If one employs a simple hydrogenic orbital for the $1s$ electrons of the helium atom, rather than a true Hartree-Fock orbital, then the integrals (47) can be evaluated in closed form. The best hydrogenic orbital is[21]

$$u_0(\vec{r})=(Z^3/\pi a_0{}^3)^{1/2}e^{-Zr/a_0},\quad Z=\tfrac{27}{16}=1.69. \tag{51}$$

Then the overlap integral (36) is

$$w(\vec{R})=(1+X+\tfrac{1}{3}X^2)\,e^{-X},\quad X=ZR/a_0, \tag{52}$$

and[22]

$$x=\frac{33\pi}{2Z^3}=10.7,\quad y=\frac{57\,637\pi}{12\,288Z^3}=3.05. \tag{53}$$

Then by (49) one has

$$n_0=n[1-0.061+O((\rho a_0{}^3)^2)]. \tag{54}$$

Thus, for the simplified ground state $|\psi_0\rangle$, 94% of the ^{4}He atoms are Bose-Einstein condensed into the zero-momentum single-atom ground state, or equivalently, the "condensate depletion" is 6%. For real liquid ^{4}He, the condensate occupation is much smaller; the data on inelastic neutron scattering suggest a condensate occupation of only a few percent. Such a large discrepancy is not surprising in view of the highly simplified nature of the state $|\psi_0\rangle$ upon which (54) is based. The important thing to note about (54) is that it does predict Bose-Einstein condensation, and that this condensation is directly related to ODLRO of the single-atom density matrix (in our notation, $\chi\neq 0$), in accordance with the general relationship[2-5] between ODLRO and Bose-Einstein condensation. The small but nonzero (6%) condensate depletion results solely from the "kinematical repulsion" of ^{4}He atoms incorporated into the state $|\psi_0\rangle$ via the exclusion principle. In a more realistic state in which the atomic motions are correlated in such a way as to minimize the energy-increasing tendency of the kinematical repulsion, the depletion would be much larger, since such correlated motions necessarily imply nonzero momentum components.

Let us now proceed with the evaluation of the momentum distribution function $n_{\vec{k}}'$ of the uncondensed (depleted) atoms. By (50) and (44)

$$n_{\vec{k}}{}' = \rho^2 a_0{}^3 \int [w^2(\vec{R}) I_{22}(\vec{R}) - 8w(\vec{R}) I_{13}(\vec{R}) + 2w(\vec{R}) I_{11}(\vec{R}) + 2I_{22}(\vec{R})] e^{-i\vec{k}\cdot\vec{R}} d^3R + O((\rho a_0{}^3)^3). \tag{55}$$

Inserting the definitions (45) of the $I_{\mu\nu}$ and the Fourier transforms $x_{\vec{k}}, y_{\vec{k}}, t_{\vec{k}}$ defined by[23]

$$w(\vec{R}) = (2\pi)^{-3} a_0{}^3 \int z_{\vec{k}}\, e^{i\vec{k}\cdot\vec{R}} d^3k, \quad w^2(\vec{R}) = (2\pi)^{-3} a_0{}^3 \int x_{\vec{k}}\, e^{i\vec{k}\cdot\vec{R}} d^3k, \quad w^3(\vec{R}) = (2\pi)^{-3} a_0{}^3 \int t_{\vec{k}}\, e^{i\vec{k}\cdot\vec{R}} d^3k, \tag{56}$$

one finds

$$\begin{aligned} n_{\vec{k}}{}' = {} & 2(\rho a_0{}^3)^2 x_{\vec{k}}{}^2 + (2\pi)^{-3} (\rho a_0{}^3)^2 a_0{}^3 \int x_{\vec{k}-\vec{k}'} x_{\vec{k}'}{}^2 d^3k' - 8(2\pi)^{-3} (\rho a_0{}^3)^2 a_0{}^3 \int z_{\vec{k}-\vec{k}'} z_{\vec{k}'} t_{\vec{k}'} d^3k' \\ & + 2(2\pi)^{-3} (\rho a_0{}^3)^2 a_0{}^3 \int z_{\vec{k}-\vec{k}'} z_{\vec{k}'}{}^2 d^3k' + O((\rho a_0{}^3)^3). \end{aligned} \tag{57}$$

Using the explicit expression (52) for the overlap integral of the hydrogenic orbital (51), one can evaluate $x_{\vec{k}}$, $y_{\vec{k}}$, and $t_{\vec{k}}$ in closed form. The resultant expressions are related to the integrals $\bar{w}_\mu(q)$, defined in Eqs. (A6), (A7), and (A9) of Ref. 11, by

$$a_0{}^3 z_{\vec{k}} = \bar{w}_1(q), \quad a_0{}^3 x_{\vec{k}} = \bar{w}_2(q), \quad a_0{}^3 t_{\vec{k}} = \bar{w}_3(q), \tag{58}$$

where $q = ka_0/Z$. The angular integrations in (57) are easily done in closed form, and the resultant one-dimensional integrals can be evaluated numerically[24] for a representative set of values of q. Such a numerical evaluation of (57) was carried out and, as a check, compared with a numerical evaluation[25] of the integrals (55). The result is plotted in Fig. 1. As expected, the ^{4}He atoms depleted from the Bose condensate are spread in $\vec{k}$ space by an amount of order $a_0{}^{-1}$ (actually $\sim 2a_0{}^{-1}$).

V. EXCITED AND IONIZED ATOMS

The kinematical repulsion of ^{4}He atoms implicit in the n-atom state $|\psi_0\rangle$ is expected to lead not only to the presence of atoms with nonzero momenta, but also to a small but nonzero number of atoms which are virtually internally excited, as well as a still smaller but nonzero number of virtually ionized atoms. In real liquid ^{4}He such virtual excitations arise not only kinematically (exclusion principle), but also dynamically. In fact, the van der Waals attraction, responsible for the fact that liquid ^{4}He is indeed a liquid, arises from electromagnetic interactions of virtually excited atoms (London dispersion force). Such effects are not included in the simplified model wave function (31), so that not much physical significance can be attached to the detailed numerical values of the populations of electronically excited atoms implied by the state (31). Nevertheless, the evaluation for the state (31) can serve as a simplified model of calculations that might eventually be carried out with more realistic many-atom states.

Recalling that the index α on n_α stands for $\alpha = (\vec{k}, \nu)$, where ν is the set of internal (electronic) quantum numbers of the ^{4}He atom, one can rewrite the first sum rule (17) in the form

$$\sum_{\vec{k}} n_{\vec{k}} + \sum_{\vec{k},\, \nu \neq 0} n_{\vec{k}\,\nu} + n_{\text{nuc}} = n. \tag{59}$$

The number of ionized atoms is equal to the number n_{nuc} of unbound nuclei.[26] n_0 is the condensate occupation and $n_{\vec{k}}$ $(\vec{k} \neq 0)$ the uncondensed ground-state occupation $n_{\vec{k}}{}'$ already evaluated. The sum of numbers of virtually excited and ionized atoms can then be obtained by subtraction. The sum of $n_{\vec{k}}$ over all $\vec{k}$ (including $\vec{k} = 0$) can be evaluated with the aid of (43) and the completeness relation for the exponential, yielding

$$\sum_{\vec{k}} n_{\vec{k}} = \Omega \rho_3(0\uparrow, 0\downarrow, 0;\, 0\uparrow, 0\downarrow, 0). \tag{60}$$

Then, recalling that $w(0) = 1$, one finds by (44)–(47)

$$\sum_{\vec{k}} n_{\vec{k}} = n[1 - 4\rho a_0{}^3 y + O((\rho a_0{}^3)^2)], \tag{61}$$

and hence by (59) the sum of numbers of virtually excited and ionized atoms is

$$\sum_{\vec{k},\, \nu \neq 0} n_{\vec{k}\,\nu} + n_{\text{nuc}} = 4\rho a_0{}^3 y n + O((\rho a_0{}^3)^2 n). \tag{62}$$

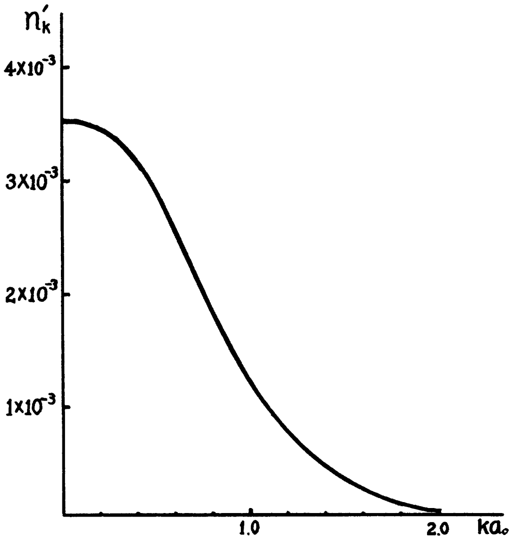

FIG. 1. Ground-state atomic momentum distribution, evaluated at liquid-^{4}He density $\rho a_0{}^3 = 0.0033$.

According to (53), the numerical value at liquid-^{4}He density ($\rho a_0{}^3 = 0.0033$) is

$$\sum_{\vec{k},\nu\neq 0} n_{\vec{k}\nu} + n_{\text{nuc}} = 0.041n. \tag{63}$$

Comparing this with (54), one sees that of the small fraction of atoms depleted from the condensate, one-third go into the momentum distribution $n_{\vec{k}}'$ ($\vec{k}\neq 0$) of unexcited atoms, and two-thirds go into virtually excited or unbound states. In a more realistic wave function one would expect not only a much larger fraction depleted from the condensate, but also a much larger number in the momentum distribution $n_{\vec{k}}'(\vec{k}\neq 0)$ than in virtually excited or ionized states.

VI. DISCUSSION

The most important result of this analysis is the unification of the viewpoint of superfluidity of liquid ^{4}He as arising from ODLRO of the two-electron one-nucleus density matrix[3,5,11] with that in which it arises from condensation of "elementary" bosons.[1-3] In our approach the operators $a_{\vec{k}}$, $a_{\vec{k}}^\dagger$ describe elementary bosons ("ideal atoms"), but nevertheless the detailed electronic structure of the atoms enters in the evaluation of $n_{\vec{k}} = \langle\psi_0|Ua_{\vec{k}}^\dagger a_{\vec{k}} U^{-1}|\psi_0\rangle$. Although the model liquid-^{4}He ground state $|\psi_0\rangle$ employed in this paper is much too crude to yield accurate numerical results, one expects these qualitative features to hold for more accurate approximations, and in fact for the true liquid-^{4}He ground state. Contrary to the assertions in Refs. 6 and 11, Bose-Einstein condensation of ^{4}He atoms is compatible with the effects of interatomic electron exchange, although these effects do cause a partial[27] depletion of the condensate.

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[12] Since the states ϕ_α are assumed to be *bound* atomic states, it would be necessary to adjoin appropriate continuum states in case ions are present.

[13] See Eq. (46) of Ref. 12, from which a factor n is missing on the right-hand side [Eq. (47) and subsequent equations are correct as written], and ρ_3 stands for D_3 defined in Eq. (41) of Ref. 12, differing from (27) in normalization.

[14] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957). The fact that the $2n$-electron projection of a BCS state is of the form (29) was pointed out by K. Nakamura, Prog. Theor. Phys. **21**, 713 (1959).

[15] We assume periodic boundary conditions in a cubical box of volume Ω.

[16] Strictly speaking, the periodic boundary conditions introduce a slight asphericity; however, this effect is utterly negligible, since the range a_0 is microscopic, whereas Ω is macroscopic.

[17] In view of the very small ratio of the electronic to nuclear mass, we approximate the center-of-mass position $R_{\text{c.m.}}$ by the nuclear position $\vec{R}$ in the phase factor; otherwise, $e^{ik\cdot R}$ would be replaced by $e^{i\vec{k}\cdot\vec{R}_{\text{c.m.}}}$.

[18] Equations (41), (42), and (B1)–(B3) of Ref. 11.

[19] Equations (43) and (44) of Ref. 11.

[20] Equation (25) of Ref. 11.

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[22] Equation (A4) of Ref. 11. The numerical value given there for y is incorrect, the correct value being $y = 3.05$; the analytical value is correct.

[23] Equation (29) of Ref. 11 and Eq. (79) of the second Ref. 6.

[24] In fact, the integrals can in principle be evaluated analytically by contour integration. However, the analytical evaluation is very complex because of the necessity of evaluating residues at high-order poles. For example, the evaluation of $\int z_{\vec{k}-\vec{k}'} z_{\vec{k}'}^2 d^3k'$ requires evaluation of residues of an expression of the form N/D, where D has eighth-order and third-order zeroes, and N is of sixth degree. The numerical evaluation involves much less work.

[25] The functions $I_{\mu\nu}$ in the integrands can in principle be evaluated analytically, and the analytical form of I_{11} is exhibited in Eq. (A10) of Ref. 11. However, it is much easier to evaluate I_{22} and I_{13} numerically, and this was done.

[26] In a more realistic calculation one would also include singly-ionized atomic states, in which case n_{nuc} would be equal to the number of *completely* (doubly) ionized ^{4}He atoms. We shall ignore such niceties here.

[27] It is conceivable that the attractive portion of the true helium-helium interatomic interaction could result in a "generalized" Bose-Einstein condensation of the type described, e.g., in Ref. 4. However, this would not invalidate the concept of a nonzero "condensed fraction."