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Proposal for Measurement of Helium-II Condensate

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A method is proposed for an experimental determination of the equilibrium condensate fraction in He^{4}II which relies only on measurement of the liquid structure factor as a function of temperature.
Existing data at 0.79 °K and 1.4 °K indicate the value to be about 10%.

Es wird ein Verfahren zur experimentellen Bestimmung des Gleichgewichtkondensationsbruchteils in He^{4}II vorgeschlagen, welches nur die Messung des Flüssigkeitsstrukturfaktors als Funktion der Temperatur erfordert.
Bestehende Meßwerte bei 0,79 °K und 1,4 °K ergeben einen Wert von ca. 10%.

Nous proposons pour la détermination expérimentale de la fraction de condensation d'équilibre dans l'He^{4}II une méthode qui ne demande que la mesure du facteur de structure du liquide en fonction de la température.
Les dates existantes pour 0.79 °K et 1.4 °K donnent une valeur d'environ 10%.

1. Introduction

London [1] first pointed to the similarities between the condensation which occurs in the ideal Bose-Einstein gas and the lambda transition in liquid He^4. Penrose [2] generalized the concept of condensation in the ideal gas to encompass the case of interacting bosons, through the introduction of a certain factorization of the first order reduced density matrix. Later Yang [3] extended this concept to include superconductivity and called it "Off-Diagonal-Long-Range-Order" (O.D.L.R.O.). This new kind of order — not of the obvious spatial kind existing, for example, in solids — also occurs in the radiation field of a laser [4], and it has recently been speculated that it may occur in certain biological systems as well [5].

In superfluid He^{4}II below the lambda transition, Penrose [2] conjectured that the first order reduced density matrix $\Omega_1(\boldsymbol{x}';\boldsymbol{x}'')$, defined by

$$\Omega_1(\boldsymbol{x}';\boldsymbol{x}'') = \mathrm{Trace}\,[\psi^\dagger(\boldsymbol{x}'')\,\psi(\boldsymbol{x}')\,\Omega(t)] \tag{1.1}$$

— where $\Omega(t)$ is the von Neumann density matrix of the system and $\psi(\psi^\dagger)$ annihilates (creates) a He^4 atom at a point — will take the factorized form:

$$\Omega_1(\boldsymbol{x}';\boldsymbol{x}'') = \Phi^*(\boldsymbol{x}'')\,\Phi(\boldsymbol{x}') + \Lambda_1(\boldsymbol{x}';\boldsymbol{x}'') \tag{1.2a}$$

where

$$\Lambda_1(\boldsymbol{x}';\boldsymbol{x}'') \xrightarrow[|\boldsymbol{x}'-\boldsymbol{x}''|\to\infty]{} 0\,. \tag{1.2b}$$

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At absolute zero ($T = 0$) the density associated with $\Lambda_1(\boldsymbol{x}; \boldsymbol{x})$ is usually called the "depletion" and is believed to account for about 90% of the total density [6], [7]. It is to be noted that unlike the case of the ideal Bose-Einstein gas the condensate density $|\Phi|^2$ is *not* the superfluid density which at $T = 0$ becomes equal to the total density.

Although the belief seems rather widespread that the appearance of Φ is of crucial physical significance, this belief is by no means universal; Landau [8], for example, never invoked the concept of "condensation", and a recent theoretical paper by Bohm and Salt [9] purports to show that no condensation occurs in Ω_1. Accordingly, direct experimental evidence of the existence of Φ would at the present time be of the utmost interest[1]; it is the purpose of this paper to give the theoretical basis of a method by which such evidence may be obtained. The idea, based on measurement of the liquid structure factor $S(\vec{k})$ by X-ray (or neutron) diffraction can be qualitatively understood by reference to a simple intuitive picture: when condensation has taken place we must not think of one He^4 atom as being in the condensate and another not; rather, each and every atom contributes both to a uniform density (the condensate), spread throughout the volume occupied by the system, *and* to localized regions of higher density — localized, of course, to within about the average inter-atomic spacing (there are always N such "lumps", where N is the total number of He^4 atoms). The diffraction pattern will derive only from the "lumps" of higher density (and not from the uniform (structure-less) background of condensate) and measurement of the relative intensity of the diffraction patterns at different temperatures will mainly reflect the degree to which the "lumps" have "melted" into the condensate as the temperature is varied. Thus above the transition temperature T_λ, we expect the positions of the peaks in the radial distribution function $g(r)$ (the Fourier transform of $S(\boldsymbol{k})$) to remain the same[2] but their amplitudes to slightly increase as T is decreased, owing to the diminution of thermal motion. For $T < T_\lambda$, however, we expect this trend to reverse, the amplitudes diminishing as the temperature is lowered, due to the lumps melting into the condensate; this reversal in trend is in fact what is observed experimentally [11]. Reference to Fig. 1 will elucidate the point; the region of interest is for $r \geqq 4.5$ Å.

In § 2 we will formulate the theory on which the above considerations are based which will lead to a simple method for the determination of the condensate fraction. § 3 will be concerned with a more detailed discussion of several points arising in § 2. In particular, in order to stress the model independence of the present method it will be useful to discuss the detailed structure of the second order reduced density matrix, Ω_2.

2. Formulation of the Method

For $r > r_0$ ($\cong 4.5$ Å) the first order reduced density matrix Ω_1 becomes equal in the case of translational and rotational invariance to the condensation density

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[1] Attempts in this direction have already been made — see Ref. [10] and references therein — but the results obtained have large systematic errors as evidenced by the fact that a 2% condensate fraction is found above T_λ, *well within* the experimental error.

[2] That is, except for a very small shift due to thermal expansion.

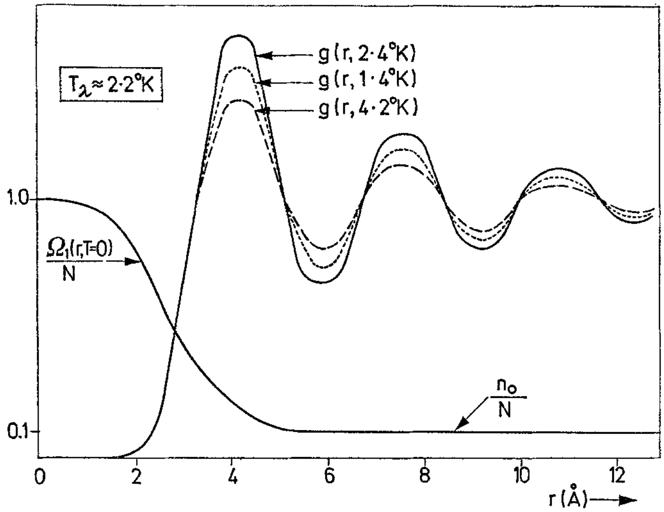

Fig. 1. The r-dependences (schematic) of the radial distribution functions $g(r, T)$, following the experimental data of Ref. [11], together with $\Omega_1(r)$ (normalized to unity), following the numerical calculations of Ref. [7]

$|\Phi|^2$ $(= n_0)$ [7] — see Fig. 1. In this limit the equilibrium pair correlation function at temperature T, $P(r, T)$ can be written — following Fröhlich [12] — in terms of the second order reduced density matrix Ω_2 as:

$$P(r, T) \equiv \Omega_2(\boldsymbol{x}, \boldsymbol{y}; \boldsymbol{x}, \boldsymbol{y} \mid T) = N^2 - [N - n_0(T)]^2 + \Lambda_2(r, T) \tag{2.1}$$

where $r = |\boldsymbol{x} - \boldsymbol{y}|$, $N (= \Omega_1(0))$ is the total number density and Λ_2 satisfies the asymptotic condition:

$$\Lambda_2(r, T) \xrightarrow[r \gg r_0]{} [N - n_0(T)]^2 . \tag{2.2}$$

$P(r, T)$ is, however, directly related to the experimentally determinable quality $g(r, T)$ — the radial distribution function — by:

$$P(r, T) = N^2 g(r, T) \tag{2.3}$$

where

$$g(r, T) \to 1 \quad \text{as} \quad r \to \infty .$$

Defining $g_N(r, T)$ by:

$$\Lambda_2(r, T) = [N - n_0(T)]^2 g_N(r, T) \tag{2.4}$$

where in order to satisfy (2.2) $\lim_{r\to\infty} g_N(r, T) = 1$, then (2.1), (2.3) and (2.4) can be written:

$$g(r, T) - 1 = \left[\frac{N - n_0(T)}{N}\right]^2 [g_N(r, T) - 1] . \tag{2.5}$$

Since the L.H.S. of (2.5) is ascertainable experimentally, determination of the condensation fraction $\frac{n_0}{N}$ as a function of T requires knowledge only of $g_N(r, T)$. It is clear from (2.5) that for $T > T_\lambda$ (where $n_0 \equiv 0$) $g(r, T) = g_N(r, T)$, so that for such temperatures[3] $g_N(r, T)$ is experimentally accessible. Now in terms of the qualitative picture presented in § 1 the X-ray (or neutron) diffraction is considered to derive solely from the non-condensate[4] (the "lumps"). Accordingly, $g_N(r, T)$ for $T < T_\lambda$ is then simply an extrapolation with T of $g_N(r, T)$ for $T > T_\lambda$, so that measurement of $g(r, T)$ for a range of T above and below T_λ permits determination of $\frac{n_0}{N}$ as a function of T for $T < T_\lambda$.

Existing experimental data [11] is available at only two temperatures above T_λ, one of which is, unfortunately, the boiling point $T_B = 4.2\,°K$; the other is $2.4\,°K$. In view of the relatively small temperature range available between T_λ and T_B ($\simeq 2°$) any significant variation of $g(r, T)$ with T is not envisaged. Making the assumption that

$$g_N(r, T) = g_N(r, T = 2.4\,°\mathrm{K}) \tag{2.6}$$

then for $T < T_\lambda$,

$$\frac{n_0(T)}{N} = 1 - \sqrt{\frac{g(r, T) - 1}{g_N(r, T = 2.4\,°\mathrm{K}) - 1}}, \tag{2.7}$$

the expression given by ourselves in Ref. [13].

From the data of Gordon *et al.* [11] it can be estimated that:

$$\frac{n_0(T = 1.4\,°\mathrm{K})}{N} \simeq 10\% \tag{2.8}$$

whilst from the data of Achter and Meyer [14] a result of a similar magnitude is obtained at 0.79 °K.

If, however, assumption (2.6) is not made but a linear extrapolation is assumed for $g_N(r, T)$ using the data at 2.4 and 4.2 °K then it is found that:

$$\frac{n_0(T = 1.4\,°\mathrm{K})}{N} \simeq 20\%\,. \tag{2.9}$$

It seems reasonable to suppose that this represents an upper limit to the ratio $n_0(T{=}1.4°\mathrm{K})$ whilst (2.8) represents a lower limit; it is gratifying to note that these values are of a magnitude comparable to those estimated (theoretically) by Penrose and Onsager [6] and McMillan [7].

The r-independence of the R.H.S. of (2.7) has been investigated firstly in terms of the positions of the zeros of the g-functions and secondly the value of the ratio at the position of the maxima and minima. It is found that the zeros coincide within 1% whilst the ratios are constant to within 10%; it should be remembered, however, that the experimental error is at least of order 5%. A further possible substantial contribution to error derives from the necessity of having to Fourier transform $S(\boldsymbol{k}, T)$ to obtain $g(r, T)$.

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[3] Through this paper it is always to be understood that $T < T_B = 4.2\,°K$, the boiling point of liquid He^4.

[4] In view of the coherency of the condensate its excitation by low energy probes (the X-rays or neutrons employed in the experimental determination of the liquid structure factor $S(\boldsymbol{k}, T)$, from which $g(r, T)$ is obtained by Fourier inversion) is not anticipated — a situation perhaps somewhat analogous to that in an electron gas in which the excitation of plasmons requires considerable energy.

Notwithstanding the relatively large error in the values of $\frac{n_0}{N}$ quoted above, it would certainly appear that the very reduction in the amplitude of the peaks in $g(r, T)$ on cooling through T_λ is convincing evidence of the existence of a condensate.

3. Discussion

In the preceding section a method was proposed whereby an experimental determination of the equilibrium condensate density could be achieved. It might appear on first sight, however, that the method relies on the particular form of Ω_2, suggested by Fröhlich, from which (2.1) then follows in an asymptotic limit. Following [12] and the references contained therein Ω_2 can be decomposed as follows[5]:

$$\begin{aligned}\Omega_2(\boldsymbol{x}',\boldsymbol{y}';\boldsymbol{x}'',\boldsymbol{y}'') = {} & [\Phi(\boldsymbol{x}')\,\Phi^*(\boldsymbol{x}'')\,\Phi(\boldsymbol{y}')\,\Phi^*(\boldsymbol{y}'') + \Phi(\boldsymbol{x}')\,\Phi^*(\boldsymbol{x}'')\,\Lambda_1(\boldsymbol{y}';\boldsymbol{y}'')\\ & + \Phi(\boldsymbol{y}')\,\Phi^*(\boldsymbol{x}'')\,\Lambda_1(\boldsymbol{x}';\boldsymbol{y}'') + \Phi(\boldsymbol{x}')\,\Phi^*(\boldsymbol{y}'')\,\Lambda_1(\boldsymbol{y}';\boldsymbol{x}'')\\ & + \Phi(\boldsymbol{y}')\,\Phi^*(\boldsymbol{y}'')\,\Lambda_1(\boldsymbol{x}';\boldsymbol{x}'') + \Lambda_1(\boldsymbol{x}';\boldsymbol{x}'')\,\Lambda_1(\boldsymbol{y}';\boldsymbol{y}'')\\ & + \Lambda_1(\boldsymbol{x}';\boldsymbol{y}'')\,\Lambda_1(\boldsymbol{y}';\boldsymbol{x}'')\\ & + \Lambda_2^{\mathrm{F}}(\boldsymbol{x}',\boldsymbol{y}';\boldsymbol{x}'',\boldsymbol{y}'')]\,S(|\boldsymbol{x}'-\boldsymbol{y}'|)\,S(|\boldsymbol{x}''-\boldsymbol{y}''|)\,.\end{aligned} \tag{3.1}$$

For our purposes it will be convenient to combine the last three terms in (3.1) and define:

$$\begin{aligned}\Lambda_2(\boldsymbol{x}',\boldsymbol{y}';\boldsymbol{x}'',\boldsymbol{y}'') \equiv {} & \Lambda_1(\boldsymbol{x}';\boldsymbol{x}'')\,\Lambda_1(\boldsymbol{y}';\boldsymbol{y}'')\\ & + \Lambda_1(\boldsymbol{x}';\boldsymbol{y}'')\,\Lambda_1(\boldsymbol{y}';\boldsymbol{x}'') + \Lambda_2^{\mathrm{F}}(\boldsymbol{x}',\boldsymbol{y}';\boldsymbol{x}'',\boldsymbol{y}'')\,.\end{aligned} \tag{3.2}$$

In the case of translational-rotational invariance the pair correlation function derived from (3.1) in the limit $\boldsymbol{x}' = \boldsymbol{x}''$; $\boldsymbol{y}' = \boldsymbol{y}''$ is a function only of $r = |\boldsymbol{x} - \boldsymbol{y}|$, whence

$$\lim_{r\to\infty} \Lambda_2(r, T) = [N - n_0(T)]^2 \qquad (\text{c.f. } \lim_{r\to\infty} \Lambda_2^{\mathrm{F}}(r) = 0)\,. \tag{3.3}$$

From the Monte Carlo calculations of McMillan [7] it can be seen that for $r > r_0$ ($\simeq 4.5$ Å), $\Lambda_1(r) \to 0$, so that r_0 can be considered to define a characteristic length associated with the non-condensate. Thus for $r > r_0$, the above form (3.1) for Ω_2 reduces, using (3.2) to the form given by (2.1). It is evident that any terms in a decomposition of Ω_2 additional to those given explicitly in (3.1) and which involve $\Lambda_1(r)$ as a factor, or alternatively have a characteristic range of order r_0 can lead to no change in the form of (2.1); *to this extent* the results of § 2 are model independent.

It is also possible to introduce a characteristic length r_1 for $\Lambda_2(r)$. For from the definition of $g(r)$ it is clear that its r-dependence reflects that of $\Lambda_2(r)$ and from the data of Ref. [11] it can be seen that $g(r)$ (and hence $\Lambda_2(r)$) approach

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[5] The S factors are functions only of the magnitude of their argument, vanishing when $|\boldsymbol{x} - \boldsymbol{y}| \to 2a_0$ and tending to unity for $|\boldsymbol{x} - \boldsymbol{y}| > 2a_0$, where a_0 is the radius of the hard core of the inter-Helium atom potential. These factors serve to "screen out" the rapidly varying repulsive core and are necessary to ensure that the pair correlation function vanishes for $|\boldsymbol{x} - \boldsymbol{y}| < 2a_0$. The possibility of more general screening factors whose arguments involve 3 space points has more recently been suggested [15].

their asymptotic constant value for $r \simeq r_1$ where $r_1 \geqq 4 r_0$; it is to be noted that the characteristic length of Λ_2 exceeds that of Λ_1.

Inspection of the various terms in the decomposition (3.1) of Ω_2 reveals that Φ and its complex conjugate always appear together in pairs. The 5 criteria given by Fröhlich [12] do not preclude, however, the possibility of terms, such as for example

$$\begin{aligned}[\Phi(\boldsymbol{x}')\,\Phi(\boldsymbol{y}'')\,C^*(\boldsymbol{y};\boldsymbol{x}'') + \Phi^*(\boldsymbol{x}'')\,\Phi^*(\boldsymbol{y}')\,C(\boldsymbol{x}';\boldsymbol{y}'')]& \\ \cdot S(|\boldsymbol{x}'-\boldsymbol{y}'|)\,S(|\boldsymbol{x}''-\boldsymbol{y}''|)&\end{aligned} \tag{3.4}$$

provided only that:

$$\lim_{|\boldsymbol{y}'-\boldsymbol{x}''|\to\infty} \begin{cases} C^*(\boldsymbol{y}';\boldsymbol{x}'') \\ C(\boldsymbol{x}';\boldsymbol{y}'') \end{cases} = 0\,. \tag{3.5}$$

Accordingly, the presence of such terms (as do in fact occur in the Bogoliubov theory of superfluidity) can not affect the validity for $r > r_0$ of Eq. (2.1)[6]. Decompositions of Ω_2 found in "pairing" theories of superfluids, which contain also the concept of a single particle condensate, again do not alter the form of (2.1) for $r > r_0$.

4. Summary

A method has been proposed for a direct experimental determination of the equilibrium condensate density in liquid He^4II which involves measurement only of the liquid structure factor $S(\boldsymbol{k}, T)$ for a range of temperatures above and below T_λ.

Owing to the experimental errors in the existing data for $S(\boldsymbol{k}, T)$ — especially in the $k \to 0$ limit — and their magnification in the subsequent Fourier analysis which is necessary in order to extract $g(r, T)$ which appears in Eq. (2.7), the value of the condensate density obtained in this paper ($\simeq 10\%$) should be considered accurate only to within a factor of two or three; the data do, however, clearly reflect the *existence* of a single particle condensate.

To obtain the temperature dependence of n_0 more accurate data will, of course, be required in a range of temperatures above T_λ and for several temperatures below.

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[6] It is in fact quite probable that the very existence of such "symmetry breaking" terms be prohibited by a sixth criterion namely that of the phase invariance of Φ, [16]. For if we accept the fact that O.D.L.R.O. introduces a new physical variable, typified say by the gradient of the phase of Φ (usually identified with the superfluid velocity field) then we must further require that the physics remain unchanged by a shift in the phase of Φ by a constant.

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