

## Temperature dependence of spatial correlations in liquid $^4\text{He}$ and the Bose-Einstein condensation

G. V. Chester

*Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York 14853*

L. Reatto

*Gruppo Nazionale di Struttura della Materia, Istituto di Fisica, Università di Milano, Milano, Italy  
and Istituto di Fisica, Università di Parma, Parma, Italy*

(Received 9 April 1980)

The basis of the equation  $[1 - n_0(T_1)]^2 = [g(r, T_1) - 1]/[g(r, T_2) - 1]$  is critically examined. Here  $T_1 \leq T_\lambda$  and  $T_2 \geq T_\lambda$ . This equation, which relates the condensate fraction  $n_0(T)$  to the radial distribution function  $g(r, T)$ , was proposed by F. W. Cummings, G. J. Hyland, and G. Rowland [Phys. Kondens. Mater. **12**, 90 (1970)]. It is shown to be based on two assumptions. It cannot be derived from the Frohlich decomposition of the two-particle density matrix.

The temperature dependence of the spatial order in liquid helium-4 below the  $\lambda$  temperature is unique: When the fluid is heated at constant density the short-range order increases. This anomalous temperature dependence ends near  $T_\lambda$  and with further increase in the temperature the behavior becomes normal; i.e., the short-range order decreases. This behavior was observed in the early measurements<sup>1</sup> of the structure factor  $S(k)$  of  $^4\text{He}$ . Recently, accurate measurements of  $S(k, T)$  have been performed by x-ray<sup>2</sup> and neutron scattering<sup>3</sup> with the specific aim of studying the temperature dependence of  $S(k, T)$  and the radial distribution function  $g(r, T)$ .

Several years ago, it was proposed<sup>4</sup> that this anomalous temperature dependence of  $g(r, T)$  is due to the existence of a Bose-Einstein condensation in the superfluid phase of  $^4\text{He}$ . At a qualitative level the origin of this behavior was attributed to the spatial delocalization of He atoms in the condensate. When  $T$  increases, the condensate fraction  $n_0(T)$  decreases, and consequently more particles can contribute to spatial order. The oscillations in  $g(r, T)$ , which reflect the short-range order, increase in amplitude until  $T = T_\lambda$ , where the condensate vanishes. The behavior then changes over to that normal for a fluid, and the increase of the thermal motion diminishes the short-range order.

Cummings, Hyland, and Rowlands<sup>4</sup> (CHR) proposed an explicit expression that relates  $n_0(T)$  to the temperature dependence of  $g(r, T)$ :

$$1 - n_0(T_1) = \{ [g(r, T_1) - 1] / [g(r, T_2) - 1] \}^{1/2}, \quad (1)$$

$$T_2 \geq T_\lambda, \quad r > l_0,$$

where  $T_1$  is a temperature below  $T_\lambda$ ,  $T_2$  is a temperature immediately above  $T_\lambda$ . The functions  $g(r, T)$  and  $g_2(r, T_2)$  are to be taken for  $r > l_0$ , where  $l_0$  is the distance at which the one-particle density matrix,  $\rho_1(r)$ , has reached its asymptotic value,  $\rho_1(l_0) \approx n_0$ .

Numerical computations<sup>5</sup> show that  $l_0 \approx 4.5 \text{ \AA}$ . If Eq. (1) is to make sense it is necessary that the right-hand side of the equation be independent of  $r$ ; this implies that  $g(r, T)$  must have the form

$$g(r, T) - 1 = f(T)F(r), \quad r > l_0, \quad (2)$$

where  $F(r)$  does not depend on  $T$ . Equation (2) implies that the zeros of  $g(r, T) - 1$  are temperature independent. The CHR formula (1) implies that  $f(T)$  depends on temperature only via  $n_0(T)$  and, moreover, has the form,

$$f(T) = [1 - n_0(T)]^2. \quad (3)$$

The recent experimental data<sup>2,3</sup> on  $S(k, T)$  have been analyzed in this way and found approximately consistent with (2). Using Eq. (1), the value of  $n_0$  has been deduced at different temperatures. By extrapolating these results to  $T = 0$  one finds (at zero pressure) that  $n_0 \approx 0.13$ ; i.e., 13% of the particles are in the condensate. This value is in reasonable agreement with exact simulation results for the Lennard-Jones model of helium-4.<sup>5</sup>

The purpose of the present paper is to discuss the theoretical basis of Eq. (1). CHR start with Frohlich's decomposition<sup>6</sup> of  $\rho_1$  and  $\rho_2$ , the one- and two-particle density matrices, respectively. From the known limits<sup>7</sup> of  $\rho_1$  and  $\rho_2$  when one or more arguments are very far from each other, these decompositions are unique and show explicitly the cluster properties of the system. The cluster functions  $\Lambda_1$  and  $\Lambda_2^F$  are defined through the equations

$$\langle r'_1 | \rho_1 | r_1 \rangle = n_0 N + \Lambda_1(r'_1 - r_1), \quad (4)$$

$$\langle r'_1 r'_2 | \rho_2 | r_1 r_2 \rangle = \langle r'_1 | \rho_1 | r_1 \rangle \langle r'_2 | \rho_1 | r_2 \rangle$$

$$+ \langle r'_1 | \rho_1 | r_2 \rangle \langle r'_2 | \rho_1 | r_1 \rangle$$

$$+ n_0^2 N^2 + \Lambda_2^F(r'_1 r'_2; r_1 r_2), \quad (5)$$

where  $N$  is the total number of particles. Here, for simplicity, we have assumed the system uniform and at rest. If the system does not have a condensate the terms explicitly dependent on  $n_0$  in (4) and (5) are absent. The functions  $\Lambda_1$  and  $\Lambda_2^F$  so defined have the cluster property; they vanish when one of the arguments is far apart from the others.

The diagonal part of  $\rho_2$  is the density-density correlation function and is proportional to  $g(r)$ :

$$\langle r_1 r_2 | \rho_2 | r_1 r_2 \rangle = N^2 g(r_1 - r_2) . \quad (6)$$

On the diagonal, Eq. (5) yields,

$$g(r_1 - r_2) = 1 - n_0^2 + (\langle r_1 | \rho_1 | r_2 \rangle)^2 / N^2 + \Lambda_2^F(r_1 r_2; r_1 r_2) / N^2 , \quad (7)$$

where we have taken into account the normalization condition  $\langle r_1 | \rho_1 | r_1 \rangle = N$ . The one-particle density matrix  $\langle r_1 | \rho_1 | r_2 \rangle$  approaches<sup>5</sup> an asymptotic limit of  $n_0 N$  and this limit is reached when  $|r_1 - r_2| \geq l \approx 4.5 \text{ \AA}$ . The two-particle correlation function  $g(r)$  has a longer range and at least up to  $r \sim 10 \text{ \AA}$  oscillates around unity. Therefore, from (7) one obtains

$$\Lambda_2^F(r_1 r_2; r_1 r_2) = N^2 [g(r_1 - r_2) - 1], \quad r > l_0 ; \quad (8)$$

i.e., the diagonal part of the cluster function  $\Lambda_2^F$  coincides with  $g - 1$  when  $r > l_0$ . We stress that this relation is *always* true, when  $n_0 \neq 0$  as well as when  $n_0 = 0$ .

The fact that  $\Lambda_1$  and  $g - 1$  have a different range is an observation that led CHR to suggest Eq. (1) for  $n_0$ . They proceed in the following way. One substitutes (4) into (5) and obtains for  $\rho_2$  an expression consisting of the sum of several terms, some of which contain  $n_0$  explicitly and some that do not. CHR introduce a new function  $\Lambda_2$  that is the sum of all the terms that do not contain explicitly  $n_0$ ,

$$\Lambda_2(r'_1 r'_2; r_1 r_2) \equiv \Lambda_2^F(r'_1 r'_2; r_1 r_2) + \Lambda_1(r'_1 - r_1) \Lambda_1(r'_2 - r_2) + \Lambda_1(r'_1 - r_2) \Lambda_1(r_1 - r'_2) . \quad (9)$$

This function does not possess the cluster property and, for instance, one gets  $\Lambda_2 = N^2(1 - n_0)^2$  when  $r'_1 = r_1, r'_2 = r_2$  with  $|r_1 - r_2| \rightarrow \infty$ . In terms of this new function  $\Lambda_2$  the diagonal part of  $\rho_2$  reads

$$\begin{aligned} \langle r_1 r_2 | \rho_2 | r_1 r_2 \rangle &= N^2 g(r_1 - r_2) \\ &= n_0(2 - n_0)N^2 + 2n_0N \Lambda_1(r_1 - r_2) \\ &\quad + \Lambda_2(r_1 r_2; r_1 r_2) . \end{aligned} \quad (10)$$

From the definition (9) the diagonal part of  $\Lambda_2$ , for  $|r_1 - r_2| > l_0$ , is related to the diagonal part of  $\Lambda_2^F$  by the simple relation

$$\Lambda_2(r_1 r_2; r_1 r_2) = N^2(1 - n_0)^2 + \Lambda_2^F(r_1 r_2; r_1 r_2) , \quad (11)$$

$$|r_1 - r_2| > l_0 ,$$

where we have taken into account that  $\Lambda_1(0) = \langle r_1 | \rho_1 | r_1 \rangle - n_0 N = N(1 - n_0)$ .

At this point CHR introduce a new correlation function,  $g_N = \Lambda_2 / N^2(1 - n_0)^2$ . Using Eqs. (8) and (11) one finds

$$\begin{aligned} g_N(r_1 - r_2) &\equiv \Lambda_2(r_1 r_2; r_1 r_2) / N^2(1 - n_0)^2 \\ &= 1 + (1 - n_0)^{-2} [g(r_1 - r_2) - 1] , \end{aligned} \quad (12)$$

$$|r_1 - r_2| > l_0 .$$

This is a rewriting of Eq. (10) when  $\Lambda_1(r_1 - r_2) = 0$ . In this way the unknown function  $g_N$  is related to the measurable pair-correlation function  $g$ . For  $T > T_\lambda$  we have  $g_N = g$  because  $n_0 = 0$ . The key assumption that CHR make is that  $g_N$  is independent of the temperature for  $T \leq T_\lambda$ . From (12) it follows that this can be true only if Eqs. (2) and (3) hold for  $g(r) - 1$ . If these equations are not obeyed then the factor  $(1 - n_0)^{-2}$ , which changes with temperature at  $T_\lambda$ , will not be canceled. If (2) and (3) hold then (12) immediately leads to (1) for  $n_0$ .

The assumption that  $g_N(r, T)$  does not change with temperature is arbitrary and *unrelated* to Frohlich's decomposition of  $\rho_2$ . In fact, Frohlich's decomposition is the unique decomposition in terms of the functions  $\Lambda_1$  and  $\Lambda_2^F$  that have the cluster property. The subsequent steps (9) and (10) simply add and subtract certain terms to  $\Lambda_2$ . These algebraic steps are certainly legitimate but are also unmotivated unless one has already a theory, or a physical argument, that tells us that the temperature dependence of  $g(r)$  is given by Eq. (2) and (3). To emphasize this point, suppose that  $g(r)$  has the form given by Eq. (2), for  $r > l_0$ , but in place of (3)  $f(T)$  is given by

$$f(T) = (1 - n_0)^{2\sigma} , \quad (13)$$

where  $\sigma$  is some constant. In this case one can repeat the steps starting from Eq. (9) but with a new function  $\bar{\Lambda}_2$ :

$$\frac{\bar{\Lambda}_2(r'_1 r'_2; r_1 r_2)}{N^2} \equiv \frac{\Lambda_2^F(r'_1 r'_2; r_1 r_2)}{N^2} + \left( \frac{\Lambda_1(r'_1 - r_1)}{N} \right)^\sigma \left( \frac{\Lambda_1(r'_2 - r_2)}{N} \right)^\sigma + \left( \frac{\Lambda_1(r'_1 - r_2)}{N} \right)^\sigma \left( \frac{\Lambda_1(r_1 - r'_2)}{N} \right)^\sigma . \quad (14)$$

In Eq. (11) the term  $(1 - n_0)^2$  is now replaced by  $(1 - n_0)^{2\sigma}$  and the assumed temperature-independent function  $\bar{g}_N$  would be defined as

$$\begin{aligned} \bar{g}_N(r_1 - r_2) &\equiv \Lambda_2(r_1 r_2; r_1 r_2) / N^2 (1 - n_0)^2 \\ &= 1 + (1 - n_0)^{-2\sigma} [g(r_1 - r_2) - 1] . \end{aligned} \quad (15)$$

Again we find that  $\bar{g}_N = g$  when  $T > T_\lambda$ . If we now assume that  $\bar{g}_N$  is independent of temperature for  $T \leq T_\lambda$  the value of  $n_0$  can be obtained from the experimental  $g(r)$  by an expression similar to (1):

$$1 - n_0(T_1) = \{ [g(r, T_1) - 1] / [g(r, T_2) - 1] \}^{1/2\sigma} , \quad (16)$$

$$T_2 \geq T_\lambda, \quad r > l_0 .$$

If theory does not provide a fundamental basis for Eq. (1) we may ask if there is a way to test its validity. In this respect, two points can be made. The first is to recognize that the positions of the zeros of  $g(r, T) - 1$  do not appear to change with  $T$ . This behavior is consistent with Eq. (1). Equation (2) also requires that the peaks of  $g(r, T) - 1$  change with  $T$  by the same factor. The best experimental data are not precise enough to permit a serious test of this requirement.

The second step is a test of the quadratic dependence, Eq. (3), of  $g(r, T) - 1$  on  $1 - n_0(T)$ . On the basis of measurements of  $S(k, T)$  this test cannot be performed because the data can be interpreted equally well on the basis of (16) with any value of  $\sigma$ . Clearly the value of  $n_0$  obtained depends on the value of  $\sigma$ ; for instance, for  $\sigma = 2$  the neutron data<sup>3</sup> give a value of 6.6% for  $n_0$  at 1.0 K instead of the value 12.7% found on the basis of (1). We conclude that from measurements of  $S(k, T)$  it is not possible to deduce the value of  $n_0$  unless one is able to justify Eq. (3), an equation that at the moment must be considered a somewhat arbitrary assumption.

Assumption (3) would receive some form of support if the qualitative picture discussed by CHR of "lumps" of higher density that melt into or out of the condensate as the temperature is varied was the only picture that could explain the observed behavior of  $g(r, T)$ . We notice that the fact that the positions of the zeros of  $g(r, T) - 1$  do not change with temperature is not at all surprising. Simple liquids like argon also have this behavior.<sup>8</sup> Along an isocore the position of the main maximum of  $S(k, T)$ , and therefore the zeros of  $g(r, T) - 1$ , is very weakly dependent on temperature. The observed change is less than 1% for a change of  $T$  of about 50% of the critical temperature.

An alternative microscopic explanation of the anomalous behavior of  $S(k, T)$  has been given by De Michelis *et al.*<sup>9</sup> This is based on an explicit model of

the density matrix for liquid helium.<sup>10</sup> From this density matrix  $g(r, T)$  is calculated using Monte Carlo techniques. This density matrix corresponds to the Landau picture of <sup>4</sup>He as a gas of noninteracting excitations, phonons and rotons, and these are represented by Feynman states. It is found, indeed, that the thermal population of roton states causes an increase of the short-range order of the fluid. This is exclusively a quantum effect. An inverted  $T$  dependence is found<sup>9</sup> for  $g(r, T)$  and its origin is quite distinct from the one suggested by CHR. These two approaches predict a completely different density dependence for  $n_0(T)$ . It is at least plausible that  $n_0$  decreases when the density increases. Numerical computations<sup>5</sup> at  $T = 0$  K indicate that  $n_0$  at solidification density is reduced from the value at equilibrium density for a factor of about  $\frac{1}{4}$ . From Eq. (1) we therefore expect that the temperature dependence of  $g(r, T)$ , for  $T \leq T_\lambda$ , will be strongly reduced at high density. On the other hand, no such reduced effect is found on the basis of the Penrose density matrix. On the contrary, the effect is enhanced at higher densities because of the decrease of the roton energy gap with increasing density, leading to an increased roton population. Thus, measurements of  $g(r, T)$  along different isocores will provide a test of the two approaches. The data of Robkoff *et al.*<sup>2</sup> along an isocore at density about 10% higher than the equilibrium density suggest no reduction in the temperature dependence of  $g(r, T)$ . An analysis of these data based on Eq. (1) would be extremely interesting.

The Penrose density matrix does not give the reversal of behavior of  $S(k, T)$  at  $T_\lambda$ . The inverted  $T$  dependence continues above  $T_\lambda$ . However, this density matrix is certainly not appropriate at and above  $T_\lambda$  because it does not contain any account of the  $\lambda$  transition and, more important, the roton excitations become heavily damped at  $T_\lambda$ . It is interesting that those modes become overdamped immediately above  $T_\lambda$ , roughly where there is the crossover between the inverted and the normal  $T$  dependence of  $S(k, T)$ .

We conclude that the relation (1) has at present no sound theoretical basis and should be regarded as an assumption. Moreover, from the measured  $S(k, T)$  at a given density there is no way to test the internal consistency of Eq. (1). The value of  $n_0$  deduced<sup>2,3</sup> from experimental data is not unique unless additional physical arguments are introduced. The observed<sup>2</sup> density dependence of  $S(k, T)$  suggests the wrong density dependence for  $n_0$  if Eq. (1) is used.

The work described herein was supported by the National Science Foundation through Grant No. DMR-77-18329 to the Theory Group of the Laboratory of Atomic and Solid State Physics at Cornell University.

- <sup>1</sup>W. L. Gordon, C. H. Shaw, and J. G. Daunt, *Phys. Chem. Solids* 5, 117 (1958); D. G. Henshaw, *Phys. Rev.* 113, 9 (1960).
- <sup>2</sup>H. N. Robkoff, D. A. Ewen, and R. B. Hallock, *Phys. Rev. Lett.* 43, 2006 (1979).
- <sup>3</sup>V. F. Sears and E. C. Svenson, *Phys. Rev. Lett.* 43, 2009 (1979).
- <sup>4</sup>F. W. Cummings, G. J. Hyland, and G. Rowlands, *Phys. Kondens. Mater.* 12, 90 (1970); G. J. Hyland, G. Rowlands, and F. W. Cummings, *Phys. Lett.* 31A, 465 (1970).
- <sup>5</sup>P. A. Whitlock, D. M. Ceperley, G. V. Chester, and M. H. Kalos, *Phys. Rev. B* 19, 5598 (1979).
- <sup>6</sup>H. Frohlic, *Phys. Kondens. Mater.* 9, 350 (1969).
- <sup>7</sup>O. Penrose and L. Onsager, *Phys. Rev.* 104, 577 (1956).
- <sup>8</sup>L. Verlet, *Phys. Rev.* 165, 201 (1968).
- <sup>9</sup>C. De Michelis, G. L. Messerin, and L. Reatto, *Phys. Lett.* 66A, 484 (1978).
- <sup>10</sup>O. Penrose, in *Proceedings of the International Conference on Low Temperature Physics*, edited by J. R. Dillinger (University of Wisconsin Press, Madison, 1958), p. 117.