# Bose-Einstein Condensation and Spatial Correlations in $^4$He

J. Mayers
*ISIS Facility, Rutherford Appleton Laboratory, OX11 0QX, United Kingdom*
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A new Monte Carlo method for calculating ground-state properties of liquid $^4$He is described. It is shown that Bose-Einstein condensation (BEC) implies delocalization of the wave function. It is shown that there is no general connection between the static structure factor and BEC. It is suggested that the observed connection in liquid $^4$He is due to the creation of spaces in the liquid structure, which are required so that the wave function can delocalize, in the presence of the hard-core interactions. It is shown that this suggestion is quantitatively consistent with observations on liquid $^4$He.

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Neutron [1,2] and x-ray [3] scattering measurements of the static structure factor $S_{\vec{q}}$ in $^4$He show highly anomalous behavior. In contrast to other liquids, where spatial correlations between the positions of different atoms increase as the temperature is lowered, spatial correlations in liquid $^4$He are reduced as the temperature of the liquid is lowered below the superfluid transition. It was suggested by Hyland *et al.* [4,5] in 1970 that the static structure factor $S^N_{\vec{q}}$ of $^4$He just above the $\lambda$ transition and the corresponding function $S^S_{\vec{q}}$ in the superfluid are related by

$$[S^S_{\vec{q}} - 1] = (1 - f)^2 [S^N_{\vec{q}} - 1], \tag{1}$$

where $f$ is the fraction of atoms occupying the Bose-Einstein condensate (BEC). At a qualitative level the origin of this behavior was attributed to the delocalization of atoms in the condensate, so that these atoms do not contribute to pair correlations.

Subsequent experimental work has suggested that Eq. (1) is at least approximately satisfied in $^4$He. Values of $f$ obtained via Eq. (1), from neutron [1,2,6] and x-ray [3] measurements of the temperature and pressure dependence of $S_{\vec{q}}$ in $^4$He, agree to within a factor of $\sim$50% with neutron Compton scattering measurements [7–9] and theoretical calculations [10–12] of $f$. However, the derivation Hyland *et al.* gave for Eq. (1) is not generally accepted. Griffin [13] has questioned the validity of their assumptions and Fetter [14] has given a counterexample to Eq. (1), based on the properties of a weakly interacting Bose gas at low temperatures. Equation (1) is not obeyed by the variational calculations of Masserini *et al.* [15]. The excitation of rotons has also been proposed as an alternative explanation for the increase in spatial correlations as the temperature is raised in the superfluid phase [16–19].

In this paper, a new approach to this problem is taken. We consider the constraints placed on the form of the many-particle Schrödinger wave function by a BEC and how these constraints will affect $S_{\vec{q}}$. It will be argued that the link between spatial correlations and $f$ in $^4$He is a geometrical consequence of the hard-core repulsion between atoms and that the apparent validity of Eq. (1) is a coincidence, due to the particular hard-core radius and number density in liquid $^4$He.

Denoting the coordinates of an arbitrarily chosen particle as $\vec{r}$ and those of the $N - 1$ other particles as $\vec{s}$, the "conditional wave function" $\psi(\vec{r} \mid \vec{s})$ is defined in terms of the $N$-particle wave function $\Psi(\vec{r}, \vec{s})$ [20,21] as

$$\psi(\vec{r} \mid \vec{s}) = \Psi(\vec{r}, \vec{s}) / \sqrt{P(\vec{s})}, \tag{2}$$

where

$$P(\vec{s}) = \int |\Psi(\vec{r}, \vec{s})|^2 \, d\vec{r}. \tag{3}$$

The essential feature of $\psi(\vec{r} \mid \vec{s})$ is that it describes the $\vec{r}$ dependence of the many-particle wave function $\Psi(\vec{r}, \vec{s})$ at fixed $\vec{s}$ and its introduction could be regarded simply as a notational device. However, as shown previously [20], in the context of this problem $\psi(\vec{r} \mid \vec{s})$ can be formally treated as the wave function of a single particle, for a given configuration $\vec{s}$ of all other particles. For example, $|\psi(\vec{r} \mid \vec{s})|^2$ is the conditional probability density in space and the conditional momentum distribution $n_{\vec{p}}(\vec{s})$ is

$$n_{\vec{p}}(\vec{s}) = \left| \int \psi(\vec{r} \mid \vec{s}) \exp(i \vec{p} \cdot \vec{r}) \, dr \right|^2. \tag{4}$$

The probability that an arbitrary particle is in a momentum state $\vec{p}$ is $n_{\vec{p}}/V$, where $V$ is the total sample volume. $n_{\vec{p}}$ is a weighted average over configurations $\vec{s}$, $n_{\vec{p}} = \int P(\vec{s}) n_{\vec{p}}(\vec{s}) \, d\vec{s}$, where $P(\vec{s})$ is the probability distribution of $\vec{s}$. One can also derive an expression for the total energy of the form $E = \int P(\vec{s}) E(\vec{s}) \, d\vec{s}$, where

$$E(\vec{s}) = \frac{1}{V} \sum_{\vec{p}} \frac{p^2}{2M} n_{\vec{p}}(\vec{s}) + \int |\psi(\vec{r} \mid \vec{s})|^2 V(\vec{r}, \vec{s}) \, d\vec{r}. \tag{5}$$

This formalism allows for a new method for Monte Carlo optimization of many-particle variational wave functions. Given functional forms of $V(\vec{r}, \vec{s})$ and $\Psi(\vec{r}, \vec{s})$, randomly generate configurations $\vec{s}$, calculate $P(\vec{s})$ via Eq. (3), $E(\vec{s})$ via Eq. (5), and $E$ as a weighted average over $\vec{s}$. Variational parameters can be optimized by minimizing $E$. Similar methods are already in use [22], but it seems possible that the method outlined here would offer an increase in

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computing efficiency, since the calculation of $E(\vec{s})$ is a one-particle problem and fast Fourier transforms can be used to calculate the kinetic energy.

In order to illustrate the use of the Monte Carlo method and also the physical content of the formalism, the simple hard-core wave function first introduced by Feynman [23] is used. This is defined to be zero if any two particles approach within a distance $2a$, corresponding to the overlap of two hard spheres of radius $a$. $\Psi(\vec{r},\vec{s})$ and hence $P(\vec{s})$ have the same value for all $\vec{r},\vec{s}$, for which no two particles are closer than $2a$. The conditional wave function $\psi(\vec{r}\,|\,\vec{s})$ [20] is zero within exclusion spheres of radius $2a$ centered at the positions $\vec{s}$ and has uniform amplitude within the rest of the volume. Configurations of $N$ nonoverlapping spheres were generated numerically within a cubic volume $V$, using a random number generator. The simulation was made on a cubic array, of side 128 pixels, with periodic boundary conditions and with each sphere having a diameter of 16 pixels. $N = 191$ was chosen so that the ratio $Na^3/V$ was the same as in $^4$He at saturated vapor pressure (SVP) and $T = 0$. ($a = 1.28$ Å and $V/N = 46.2$ Å.) $\psi(\vec{r}\,|\,\vec{s})$ was generated by setting all pixels outside the exclusion spheres of radius $2a$ to the same value, which was determined by numerical normalization of $\psi(\vec{r}\,|\,\vec{s})$. The momentum distribution $n_{\vec{p}}(\vec{s})$ and the condensate fraction $f(\vec{s}) = n_0(\vec{s})/V$ were calculated from Eq. (4). [In fact, it can be shown [20] that for this model, the calculation of $f(\vec{s})$ reduces to the conceptually simple geometrical problem of finding the fraction of the total volume, which could be occupied by the center of the particle of interest, without hard-core overlap with other spheres centered at positions $\vec{s}$.]

The results of 20 000 simulations, with different randomly generated $\vec{s}$, were binned as a function of $f(\vec{s})$ and are shown in Fig. 1. The mean value obtained for $f(\vec{s})$ is $f = 0.085$, very close to the value ~0.08 obtained by Penrose and Onsager [24] with the same model but by an entirely different method of calculation. The peak shape in Fig. 1 becomes closely Gaussian at large $N$, with a width proportional to $1/\sqrt{N}$ and in a macroscopic system, the condensate fraction will be very precisely defined and independent of the precise microscopic configuration $\vec{s}$. Figure 2 shows the calculated longitudinal momentum distribution $J(u)$.

$$J(u) = \int n_p(u, p_y, p_z)\, dp_x dp_y\,. \tag{6}$$

Comparing with the calculations of Ceperley [25], shown as the dashed line, shows that this crude model gives a reasonably good description of both the condensate fraction and $n_{\vec{p}}$. The general conditions imposed upon the wave function by the presence of BEC can be understood by using the Wiener-Kintchine theorem [26] and expressing $n_{\vec{p}}(\vec{s})$ as

$$n_{\vec{p}}(\vec{s}) = \int A(\vec{r}\,|\,\vec{s}) \exp(i\vec{p}\cdot\vec{r})\, dr\,, \tag{7}$$

where $A(\vec{r}\,|\,\vec{s})$ is the autocorrelation function of $\psi(\vec{r}\,|\,\vec{s})$,

$$A(\vec{r}\,|\,\vec{s}) = \int \psi(\vec{r}\,'\,|\,\vec{s})\psi^*(\vec{r} + r'\,|\,\vec{s})\, d\vec{r}\,'. \tag{8}$$

The inverse relation to (7) is

$$A(\vec{r}\,|\,\vec{s}) = \frac{1}{V}\sum_{\vec{p}} n_{\vec{p}}(\vec{s}) \exp(-i\vec{p}\cdot\vec{r})\,. \tag{9}$$

When a BEC is present, then on average $n_0(\vec{s})/V = f$ and $A(\vec{r}\,|\,\vec{s}) \sim f \neq 0$ as $|\vec{r}| \to \infty$. This has two implications for the form of $\psi(\vec{r}\,|\,\vec{s})$ [27], in the limit $V \to \infty$.

(1) $\psi(\vec{r}\,|\,\vec{s})$ is *delocalized* and must extend with nonzero amplitude over length scales $\sim\sqrt[3]{V}$. For example, if $\psi(\vec{r}\,|\,\vec{s}) = 0$ outside any sphere of radius $L$, then $A(\vec{r}\,|\,\vec{s}) = 0$, for $|\vec{r}| > 2L$.

(2) $\psi(\vec{r}\,|\,\vec{s})$ must be *phase coherent* over length scales $\sim\sqrt[3]{V}$. For example, if the phases of $\psi(\vec{r}\,'\,|\,\vec{s})$ and $\psi(\vec{r}\,' + \vec{r}\,|\,\vec{s})$ are uncorrelated for $|\vec{r}| > L$, then the product $\psi(\vec{r}\,'\,|\,\vec{s})\psi^*(\vec{r}\,' + \vec{r}\,|\,\vec{s})$ has random phase and the integral in Eq. (8) averages to zero for $|\vec{r}| > L$. The wave function used to generate Figs. 1 and 2 satisfies both these

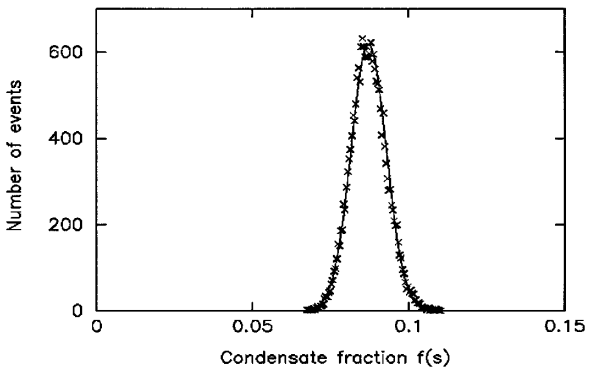


FIG. 1. The distribution of $f(\vec{s})$ values calculated for 191 spheres, with a sample of 20 000 simulations. The solid line is a Gaussian with the same mean and second moment as the data.

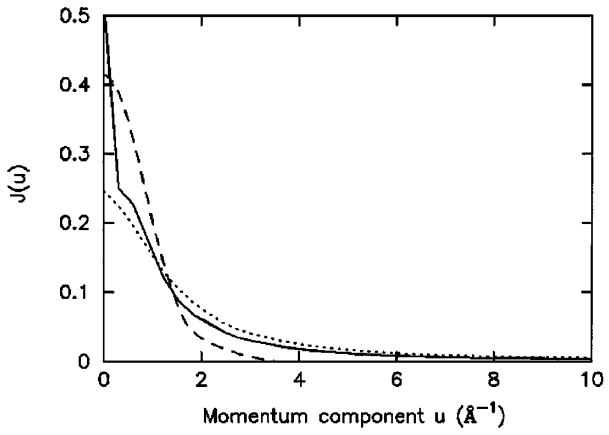


FIG. 2. The solid line shows the $J(u)$ calculated from the hard-core model. The sharp central peak is the condensate peak. The dashed line shows the noncondensate component of $J(u)$, calculated by the path integral Monte Carlo method [25]. The dotted line was calculated from a phase incoherent wave function, as described in the text.

conditions and hence produces a sharp condensate peak at $\vec{p} = 0$. It is worth noting that these two conditions imply that angular momentum must be quantized on macroscopic length scales and hence that BEC implies superfluidity [20].

$S_{\vec{q}}$ can also be expressed in terms of the many-particle wave functions $\Psi_i(\vec{r}_N)$:

$$S_{\vec{q}} = \frac{1}{N} \sum_{m,n} \sum_i B_i \int |\Psi_i(\vec{r}_N)|^2 \exp[i\vec{q} \cdot (\vec{r}_m - \vec{r}_n)]\, d\vec{r}_N \,, \tag{10}$$

where $B_i$ is the thermal probability that state $i$ is occupied and $\vec{r}_N$ denotes the coordinates of the $N$ particles. It can be seen from Eq. (10) that $S_{\vec{q}}$ depends only on $|\Psi(\vec{r}_N)|^2$ and hence is completely insensitive to the phase of $\Psi(\vec{r}_N)$. It immediately follows that there is no general connection between $S_{\vec{q}}$ and $f$, since $f$ is sensitive to the phase of $\Psi(\vec{r}_N)$, whereas $S_{\vec{q}}$ is not. In fact, from a $\Psi(\vec{r}_N)$ which gives a finite condensate fraction $f$, one can construct a class of wave functions $\Psi_e(\vec{r}_N)$, which give no BEC, but precisely the same $S_{\vec{q}}$. This can be achieved by the prescription

$$\Psi_e(\vec{r}_N) = \prod_{n=1}^{N} \exp[i\phi(\vec{r}_n)]\Psi(\vec{r}_N) \,, \tag{11}$$

where $\phi(\vec{r})$ is any function such that $\exp[i\phi(\vec{r})]$ and $\exp[i\phi(\vec{r}')]$ have uncorrelated phases for $|\vec{r} - \vec{r}'| \to \infty$. Since $|\Psi(\vec{r}_N)|^2 = |\Psi_e(\vec{r}_N)|^2$, $\Psi(\vec{r}_N)$ and $\Psi_e(\vec{r}_N)$ give the same $S_{\vec{q}}$. However, it follows from Eq. (2) that, apart from a factor independent of $\vec{r}$, the conditional wave function corresponding to $\Psi_e(\vec{r}_N)$ is

$$\psi_e(\vec{r} \,|\, \vec{s}) = \exp[i\phi(\vec{r})]\psi(\vec{r} \,|\, \vec{s}) \,. \tag{12}$$

From the definition of $\phi(\vec{r})$, it follows that $\psi_e(\vec{r} \,|\, \vec{s})$ does not have long range phase coherence in $\vec{r}$ for any $\vec{s}$ and $\Psi_e(\vec{r}_N)$ violates condition (2) for BEC. Hence $\Psi_e(\vec{r}_N)$ gives no BEC. This is illustrated in Fig. 2, where the dotted line was generated by randomly stepping the phase of the wave function used to generate the solid line, by $\pm 2\pi/m$ in adjacent pixels along the $x$ axis, with $m = 10$. The phase undergoes a random walk in one dimension and there is no phase correlation between points separated by more than $\sim m^2 = 100$ pixels. Thus there is no sharp condensate peak.

In order to show how a link between $S_{\vec{q}}$ and $f$ may arise in $^4$He, consider a modification of the hard sphere model, which was used to generate Figs. 1 and 2. Rather than being randomly positioned within the volume $V$, the $N$ spheres were positioned on the sites of a regular lattice. For each simulation there were $N_S = N/(1 - c)$ lattice sites, with a fraction $c$ of random vacancies, so that $N$ sites were occupied. The lattice constant $d$ was scaled $\propto \sqrt[3]{1 - c}$, so that $Na^3/V$ was independent of $c$ and equal to the value in liquid $^4$He at SVP. Three different cases were considered: a simple cubic lattice, a body centered cubic (bcc) lattice, and a face centered cubic (fcc) lattice. As in the previous model, $\psi(\vec{r} \,|\, \vec{s})$ had uniform amplitude within the volume outside the hard-core exclusion spheres of radius $2a$, centered at each occupied lattice site. The results of numerical calculations of $f$, calculated from Eq. (3), as a function of vacancy concentration $c$ are shown in Fig. 3. For bcc and fcc lattices $f \to 0$ as $c \to 0$, since the hard-core exclusion spheres overlap to fill all of space. For these structures there is no BEC, unless some lattice sites are unoccupied.

It is a simple matter to calculate $S_{\vec{q}} - 1$ as a function of $c$ for this model. The Bragg [28] scattering from a lattice of $N_S$ sites, a fraction $c$ of which are vacant, is proportional to $N_S(1 - c)^2$. Since $N_S = N/(1 - c)$, it follows that the Bragg intensity is proportional to $1 - c$. The lost intensity appears as a constant incoherent background, which makes no contribution to $S_{\vec{q}} - 1$. It follows that for this model $S_{\vec{q}} - 1 \propto 1 - c$. If the density and hard sphere radius are such that the condition

$$f \approx 1 - \sqrt{1 - c} \tag{13}$$

is satisfied, then $S_{\vec{q}} - 1 \propto (1 - f)^2$ in agreement with Eq. (1). Equation (13) is shown as the solid line in Fig. 3. It can be seen that the calculations for all three lattices lie close to this prediction, at the density and hard-core radius of liquid $^4$He. However, the relationship between $c$ and $f$ is very sensitive to the ratio of the lattice spacing $d$ and the hard sphere diameter $2a$. Also shown in Fig. 3 are the calculated ratios between $c$ and $f$ for three different values of the ratio $2a/d$ in a bcc lattice. It can be seen that changing this ratio by only $\sim$10% moves the calculations well away from the predictions of Eq. (1). The pressure dependence of the link between $c$ and $f$ could be used to test the mechanism proposed here. The triangles in Fig. 3 were calculated for a density corresponding to liquid $^4$He at 25 bars.

The model clearly has many inadequacies, the most obvious being that in liquid $^4$He, there is no long range order. However, the geometrical arguments limiting the size

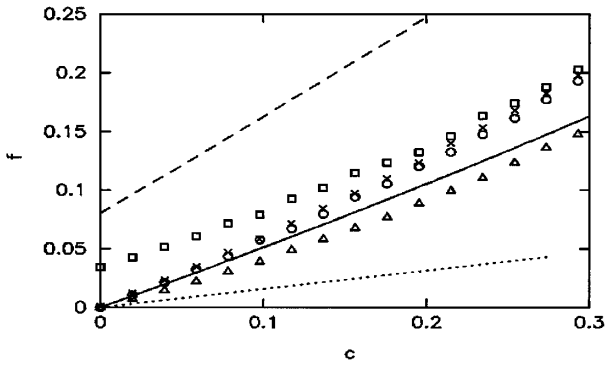


FIG. 3. The values of $f$ calculated numerically as a function of vacancy concentration $c$. The solid line is the prediction of Eq. (13). Calculations with a hard-core radius of $2a = 2.56$ Å and a density $V/N = 46.2$ Å$^3$; ○: fcc lattice; ×: bcc lattice; □: simple cubic lattice. △: calculation for a bcc lattice at $V/N = 38.9$ Å$^3$, corresponding to a pressure of 25 bars. The dashed line was calculated for a bcc lattice, with $2a/d = 0.5$ and the dotted line for $2a/d = 0.7$. At the density of $^4$He at SVP, $2a/d = 0.566$.

of the condensate factor depend only upon the local configurations of atoms, surrounding a given atom. Providing that this is approximately fcc or bcc, the limitations placed on the size of $f$ by the structure and hard sphere interaction will still apply. Furthermore, the random removal of a fraction $c$ of atoms will reduce $S_{\vec{q}} - 1$ by the factor $(1 - c)^2$ whether or not long range order is present. Another approximation of the model is that $\psi(\vec{r} \mid \vec{s})$ is a constant in the regions of space outside the hard-core exclusion spheres. It can be shown that $f$ will be *reduced* if $\psi(\vec{r} \mid \vec{s})$ decreases more smoothly outside the exclusion spheres. Thus in a more realistic model, the atoms must be arranged so that *at least* a fraction $f$ of the total volume lies outside the hard-core exclusion spheres.

Probably the most serious approximation of the lattice model is that the vacancies in the structure are produced by randomly removing atoms. This implies that [29] $S_{\vec{q}}$ contains a constant diffuse background of intensity $c$, independent of $q$ and that $S_{\vec{q}} \neq 0$ as $q \rightarrow 0$, whereas in $^4$He the Feynman relation [30] is obeyed. The model also predicts that the positions of peaks and troughs in $S_{\vec{q}}$ will shift by $\sim 2\%$, due to the lattice contraction required to keep $N/V$ constant and this is not observed experimentally. Both these undesirable features can be eliminated by the assumption that, rather than random vacancies appearing in a fixed structure, the structure distorts to create the space necessary for BEC [29].

To summarize, in liquid $^4$He, the number density and atomic hard-core diameter is such that for some atomic arrangements (e.g., bcc or fcc local ordering), delocalization cannot occur, due to a lack of space. It has been suggested that the observed loss of spatial correlations in $^4$He is due to a rearrangement of atoms, to create the necessary spaces for delocalization to occur. The lattice model shows that such a mechanism is quantitatively consistent with observations on $^4$He. However, the link between $S_{\vec{q}}$ and $f$ is very sensitive to the hard-core radius and number density and the numerical agreement of Eq. (1) with experiment in $^4$He is probably coincidental.


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