Bose-Einstein Condensation in solid $^4$He

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We present neutron scattering measurements of the atomic momentum distribution, $n(k)$, in solid helium under a pressure $p = 41$ bars and at temperatures between 80 mK and 500 mK. The aim is to determine whether there is Bose-Einstein condensation (BEC) below the critical temperature, $T_c = 200$ mK where a superfluid density has been observed. Assuming BEC appears as a macroscopic occupation of the $k = 0$ state below $T_c$, we find a condensate fraction of $n_0 = (-0.10 \pm 1.20)\%$ at $T = 80$ mK and $n_0 = (0.08 \pm 0.78)\%$ at $T = 120$ mK, consistent with zero. The shape of $n(k)$ also does not change on crossing $T_c$ within measurement precision.

In 2004, Kim and Chan [1, 2] reported the spectacular observation of a superfluid density in solid helium below a critical temperature, $T_c$. The superfluid density, $\rho_S$, is observed as a non-classical rotational inertia (NCRI) in a torsional oscillator (TO) containing solid helium. The percent of solid that has a NCRI and is decoupled from the rest of the solid was obtained in commercial grade purity $^4$He which contains typically 0.3 ppm of $^3$He, where $T_c = 200$ mK. All other impurities are frozen out. A $\rho_S$ was observed in both bulk solid helium [2] and in solid confined in porous media (Vycor) [1]. The magnitude of $\rho_S$ varies somewhat from solid sample to solid sample [3]. A superfluid density was observed in solids at pressures between $p \sim 25$ bars near the melting line and 135 bars with $\rho_S$ taking its maximum value of 1.5 % at $p \sim 50$ bars.

This remarkable result has been confirmed in independent TO measurements [4, 5, 6]. Rittner and Reppy [7] find that $\rho_S$ can be significantly reduced by annealing the solids near their melting temperatures with $\rho_S$ reduced to zero in some cases. Similarly, Shirahama and coworkers [4] report a reduction in $\rho_S$ of up to 50 % by annealing. Macroscopic superflow was not observed in helium in Vycor [7] and bulk helium [8]. However, Sasaki et al. [9] have observed macroscopic superflow in those solids which contain grain boundaries that extend across the solid. This unexpected result suggests that there is indeed superflow and that it is long or associated with grain boundaries. Superflow related to grain boundaries [9], the variation of $\rho_S$ from sample to sample [3] and the reduction of $\rho_S$ by annealing [4, 6] suggest that a superfluid density may be associated with macroscopic defects that extend across or whose impact extends across the whole solid.

In liquid helium, Bose-Einstein condensation (BEC) and superfluidity are observed together. Indeed superflow can be shown to follow from BEC [10, 11]. It can also be shown to arise from long range atomic exchanges [12]. In the liquid, both in bulk [13] and in Vycor [15], BEC is observed as a macroscopic occupation of the $k = 0$ state in $n(k)$, expected for a translationally invariant system. Solid helium is also translationally invariant, as is a gas of vacancies in the solid. Thus, for certain models of superflow we anticipate that BEC will appear as a macroscopic occupation of $k = 0$ in $n(k)$. In this context we look for an enhancement of $n(k)$ at $k \sim 0$ below $T_c$. We also look for a change in shape of $n(k)$ below $T_c$. The central result is that we observe no increase in $n(k)$ at $k \sim 0$ nor any change in shape of $n(k)$ as the temperature is lowered below $T_c$.

In 1969, Andreev and Lifshitz [16] proposed that helium could be a supersolid if the solid contained vacant sites in the ground state. Essentially, the ground state, zero point vacancies could form a Bose gas at $T \sim 0$ K in which the Bose-Einstein condensate fraction, $n_0$, and $\rho_S$ are both approximately 100 %. While thermally activated vacancies have been observed [17], ground state vacancies have not. Chester [18] proposed that superflow in solid helium may be possible because the solid is well described by fluid like wave functions that support superflow. Leggett [19] examined superflow via long range exchanges of atoms within a perfect helium solid and found superflow possible but that $\rho_S$ would be small, $\rho_S \sim 0.01 \%$.

Recent accurate path integral Monte Carlo calculations [20] find that $\rho_S$ arising from long range atomic exchanges in a perfect crystal will be unobservably small. Similarly, BEC in bulk perfect crystals is predicted to be very small [21, 22]. Zero point vacancies in the ground state are predicted to be unstable [21]. Essentially, vacancies migrate to a surface or coalesce to create a surface and leave the crystal. However, if solid helium is held in an amorphous rather the equilibrium crystal state, both $\rho_S$ and $n_0$ take significant values [21]. With these predictions, it is interesting to search for BEC where $\rho_S$ is observed.

The solid $^4$He investigated in this experiment was
The block was observed at filling an Oxford Instruments Kelvinox VT dilution refrigerator.

Commercial purity $^4$He (0.3 ppm $^3$He) was introduced into a cylindrical Al sample cell of 20 mm diameter and 62 mm height at a temperature $T \approx 3$ K to a pressure of $p \approx 70$ bars. At constant $p$, the temperature was reduced using an Oxford Instruments Kelvinox VT dilution refrigerator until solid formed in the capillary and blocked the cell. The block was observed at filling $p = (69.8 \pm 0.2)$ bars and $T = (2.79 \pm 0.02)$ K, which corresponds to liquid on the melting line at a molar volume $V_m = (20.01 \pm 0.02)$ cm$^3$/mol $^{22}$.

The mean square atomic momentum, $\sigma_2 = \langle k^2 \rangle$ along an axis and the atomic kinetic energy $K = (\lambda^2/2)\sigma_2$ where $\lambda = 12.12$ K $\text{Å}^2$ of solid helium at $V = 20.01$ cm$^3$/mole versus temperature.

Specifically, at $hQ \to \infty$, where the Impulse Approximation (IA) is valid, the energy transfer $\hbar\omega$ in $S(Q, \omega)$ is Doppler broadened by the atomic momentum distribution $n(k)$. In this limit, it is convenient to express $\omega$ in terms of the ‘$y$ scaling’ wave vector variable, $y = (\omega - \omega_R)/v_R$ where $\omega_R = hQ^2/2m$ and $v_R = hQ/m$, and to present the neutron inelastic data as $J(Q, y) = v_R S(Q, \omega)$. Including final state (FS) effects which are small but not negligible at the $Q$ values investigated here,

$$J(Q, y) = \int dq R(Q, y - y')J_{IA}(y') \tag{1}$$

where $R(Q, y)$ is the FS broadening function and

$$J_{IA}(y) = \int dq n(k)\delta(k - Q - y) = n_Q(y) \tag{2}$$

is the IA to $J(Q, y)$. Specifically, $J_{IA}(y)$ is $n(k)$ projected along $Q$ denoted the longitudinal momentum distribution $^{25}$.

Fig. 1 shows the observed $J(Q, y)$ at wavevectors $Q = 26$ $\text{Å}^{-1}$ and temperatures $T = 120$ mK and $T = 300$ mK. The observed $J(Q, y)$ includes the MARI instrument resolution function which is shown separately as a dotted line in Fig. 1. The solid line is a fit of a model to the data as described below. We were able to determine at most two free parameters in model fits to the data.

To obtain a condensate fraction, we assumed a model $n(k)$ of the form,

$$n(k) = n_0\delta(k) + (1 - n_0)n^*(k) \tag{3}$$

where $n^*(k)$ is the momentum distribution of the atoms above the condensate in the $k > 0$ states. To proceed, we
We first fit the model \( n(k) \) to the data at 500, 300, 120 and 80 mK assuming \( n_0 = 0 \) at all temperatures. The \( n_0 \) is expected to be zero at 300 and 500 mK. The resulting values of \( \overline{\sigma}_2 \) and the associated atomic kinetic energy \( (K = (3h^2/2m)\overline{\sigma}_2) \) are shown in Fig. 2. The resulting \( \overline{\sigma}_2 \) and \( K \) decrease somewhat with decreasing temperature. In a recent measurement, Adams et al. [14] find \( K \) independent of temperature within precision.

If superflow is associated with defects in the solid such as vacancies, we anticipate that BEC is similarly associated with these defects, perhaps in a vacancy gas. In this event, the atomic kinetic energy, \( K \), of the majority of the atoms may be largely unaffected by the BEC in the defects. To obtain \( n_0 = 80 \text{ mK} \) and 120 mK within this picture we kept \( \overline{\sigma}_2 \) fixed at the values obtained above and refitted model [3] with \( n_0 \) as a free parameter. The fitted values of \( n_0 \) are shown in Fig. 3. The variation of \( n_0 \) with \( Q \) reflects the statistical error in \( n_0 \). The mean values are \( n_0 = (-0.1 \pm 1.2) \% \) at 80 mK and \( n_0 = (0.08 \pm 0.78) \% \) at 120 mK.

If, in contrast, the superflow and BEC lie within the bulk of the solid, we anticipate, as in liquid \(^4\)He, that the atomic kinetic energy, \( K \), will decrease below \( T_c \) as a result of BEC. The observed decrease in \( K \) below \( T_c \) has been used to estimate \( n_0 \) in liquid \(^4\)He [26, 27]. To address this case, we kept \( \overline{\sigma}_2 \) (kinetic energy arising from \( n^*(k) \)), constant at the value obtained at 300 mK and refitted model [3] at 80 mK and 120 mK to obtain \( n_0 \). The resulting values are \( n_0 = (0.8 \pm 1.2) \% \) at 80 mK and \( n_0 = (0.76 \pm 0.77) \% \) at 120 mK. This method assumes that all the drop in \( K \) below \( T_c = 200 \text{ mK} \) arises from BEC. To normalize these \( n_0 \) values for changes in \( K \) from other sources, we also determined \( n_0 \) at 300 mK. That is, we kept the \( \overline{\sigma}_2 \) fixed at its 500 mK value and refitted the model to determine \( n_0 \) at 300 mK giving \( n_0 = (0.63 \pm 0.77) \% \). Since \( n_0 \) at 300 mK is zero, we expect the \( n_0 \) values below \( T_c \) to be overestimated by approximately 0.6 \%. Normalizing for this overestimate, we arrive at \( n_0 \approx (0.2 \pm 1.2) \% \) at 80 mK and \( n_0 \approx (0.1 \pm 0.8) \% \) at 120 mK. If we try to determine both \( \overline{\sigma}_2 \) and \( n_0 \) simultaneously, we get essentially the same values as before, with only larger error bars; e.g. \( n_0 = (0.74 \pm 1.01) \% \) at \( T = 120 \text{ mK} \). Thus all methods give similar values of \( n_0 \).

To investigate a possible change in shape of \( J(Q,y) \) or \( n(k) \) on crossing \( T_c = 200 \text{ mK} \), we fit the additive approach \( (AA) \) [28] to the data. In this model to lowest order, \( J(Q,y) \) is [13, 28]

\[
J(Q,y) = \left[ 1 - \frac{\overline{\sigma}_4}{2\gamma_2^{1/2}} \left( x - \frac{x^3}{3} \right) + \frac{\overline{\sigma}_4}{8\gamma_2} \left( 1 - 2x^2 + \frac{x^4}{3} \right) \right] J_G(x) \tag{4}
\]

where \( J_G = \frac{1}{\sqrt{2\pi\overline{\sigma}_2}} \exp(-x^2/2) \) and \( x = y/\overline{\sigma}_2^{1/2} \). The second term in (4) is the leading FS term and the third term is the leading correction to a Gaussian \( n(k) \) plus a FS term. The fitting parameters are \( \overline{\sigma}_2, \overline{\sigma}_3 = \overline{\sigma}_3/\lambda Q, \) and \( \overline{\sigma}_4 = \overline{\sigma}_4 + \overline{\sigma}_4/\lambda Q \). Previously we found \( \overline{\sigma}_4 = 0 \) [13, 14, 23]. The three remaining parameters are \( \overline{\sigma}_2, \overline{\sigma}_3, \) and \( \overline{\sigma}_4 \). The kurtosis of \( n(k) \) is \( \delta = \frac{\overline{\sigma}_4}{\overline{\sigma}_2^2} \).
This also predicts $n_0 \sim 1.5 \%$. This large value of $n_0$ and this simple model appears to be excluded by our observed values, of $n_0 = (-0.10 \pm 1.20) \%$. In contrast, the $n_0$ values within bulk solid helium including vacancies or in extended amorphous regions, whether in equilibrium or not, are consistent with our observed value.

In summary, we have determined the BEC condensate fraction in commercial grade purity solid helium at pressure 41 bars and molar volume 20.01 cm$^3$/mole using inelastic neutron scattering. We find a condensate fraction $n_0 = (0.10 \pm 1.20) \%$ below $T_c = 200$ mK consistent with zero. We also find no change in the shape of the atomic momentum distribution on crossing $T_c$.

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