The Quest for Bose-Einstein Condensation in Solid $^4$He

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Abstract  Ever since the seminal torsional oscillator (TO) measurements of Kim and Chan which suggested the existence of a phase transition in solid $^4$He, from normal to a ’supersolid’ state below a critical temperature $T_c = 200$ mK, there has been an unprecedented amount of excitement and research activity aimed at better understanding this phase. Despite much work, this remarkable phase has yet to be independently confirmed by conventional scattering techniques, such as neutron scattering. We have carried out a series of neutron scattering measurements, which we here review, aimed at observing Bose-Einstein condensation (BEC) in solid $^4$He at temperatures below $T_c$. In bulk liquid $^4$He, the appearance of BEC below $T_s$ signals the onset of superfluidity. The observation of a condensate fraction in the solid would provide an unambiguous confirmation for ’supersolidity’. Although, our measurements have not yet revealed a non-zero condensate fraction or algebraic off diagonal long-range order $n_0$ in solid $^4$He down to 65 mK, i.e. $n_0 = (0 \pm 0.3)\%$, our search for BEC and its corollaries continues with improved instrumentation.

Keywords  BEC of excitations · Supersolids · Glasses and defects
1 Introduction

The apparent decoupling of a small fraction of $^4$He atoms from the remainder of the atoms in solid $^4$He observed in torsional oscillator (TO) measurements continues to be the source of much debate in the condensed matter community. The observation was first reported by Kim and Chan [1, 2] below a critical temperature $T_c \sim 200$ mK and later reproduced and confirmed by several other researchers [3–5] around the world. This non-classical rotational inertia, denoted NCRI, has been interpreted by Kim and Chan as a signature of supersolid behavior, remarkably extending superflow to solids. This discovery has revived the old problem of superfluidity and the hypothetical supersolidity, giving birth to a whole new field of physics (new PACS Index 67.80.bd, supersolid $^4$He).

The fraction of superflow, $\rho_s/\rho$, reported changes dramatically, from 0.015% to 20%, depending on the sample growth technique, on subsequent annealing, and on other factors [6–9]. This suggests that NCRI may be associated with some kind of defects such as dislocations, grain boundaries, point defects, glassy regions, or surfaces. Indeed, Monte Carlo calculations predict that for perfect crystals of solid $^4$He both the $\rho_s/\rho$ and the Bose-Einstein condensation (BEC) fraction, $n_0$, are unobservably small [10–13]. However, observable values of $\rho_s/\rho$ and the condensate fractions $n_0$ (~0.3–0.5%) have been calculated for glassy (amorphous) solid helium [11] and in solid $^4$He containing vacancies [12, 13]. Amorphous regions in bulk solid helium [14] and completely amorphous solid helium in porous media [15, 16] have now been experimentally observed. Interestingly, Rittner and Reppy [3] found large superfluid fractions in solid samples that have a large surface area to volume ratio $S/V$ with $\rho_s/\rho$ as large as 20% at $S/V = 150$ cm$^{-1}$. Unfortunately, these large $\rho_s/\rho$ values are not universally observed and may arise from rapid cooling of thin samples [17].

Surprisingly, the sheer modulus $\mu$ of solid $^4$He, a measure of the rigidity, shows [8] an unexpected increase below $T_c$. The increases in $\mu$ and in $\rho_s/\rho$ have the same dependence on temperature and on $^3$He concentration, suggesting a common physical origin. However, recent measurements [18] show that $\mu$ increases in hcp crystals only (both $^4$He and $^3$He) while an NCRI is observed in $^4$He only, both bcc and hcp. An unexplained excess in the heat capacity [19, 20] that peaks at $T_c$ and bulk mass flow in $^4$He solids near the melting line [21] have been reported. All these suggest that NCRI may be a property that is unique to $^4$He.

As in the case of superfluid $^4$He, an observation of BEC below $T_c$ in solid $^4$He would be an unambiguous verification that the observed NCRI indeed arises from superflow. In bulk three-dimensional systems, superflow is a consequence of BEC. In two-dimensional bulk systems, the onset of superflow is associated with the onset of order [22] that can be observed [23] in a similar way as BEC. We here review our recent neutron scattering efforts aimed at observing Bose-Einstein condensation (BEC) in solid $^4$He at temperatures below $T_c$.

2 Inelastic Neutron Scattering

Neutron scattering is unarguably an invaluable tool to study structural and dynamical properties in condensed matter physics. This is because thermal neutrons are such
that their energies, are of the order of atomic excitation energies (few meVs to a few eVs) and their wavelengths match closely interatomic distances (0.05–20 Å). The particular neutron scattering technique used to measure momentum distribution of atoms in liquid and solid helium is known as deep inelastic scattering (DINS). This technique probes the dynamics of single $^4$He atoms in the atto-seconds ($\sim 10^{-18}$ s) time scale and becomes possible when the momentum and energy transferred to the atoms are significantly large compared to the atomic collective excitations wavevectors and energies [24–27]. In this limit, high energy incoming neutrons (i.e. having short wavelength) interact with single atomic nuclei in the material and transfer ‘impulses’ to individual atoms. This is known as the impulse approximation limit. At high $Q$, the energy transferred to the sample, $\hbar \omega$, is large compared to the collective excitations energies in the material and the scattering time is very brief. As a result, the struck atoms recoils somewhat independently of its neighbors and we expect the observed scattering intensity to be well approximated by the incoherent dynamic structure factor $S(Q, \omega)$. In liquid $^4$He, this is a good approximation for $Q \geq 10$ Å$^{-1}$. In the ‘Impulse Approximation’ limit (IA), i.e. $Q \to \infty$, the scattering function reduces to

$$S_{IA}(Q, \omega) = \int dp n(p) \delta\left(\omega - \omega_R - \frac{pQ}{m}\right)$$  \hspace{1cm} (1)

where $\omega_R = \hbar Q^2/2m$ is the free atom recoil frequency. Here, it becomes convenient to express $\omega$ in terms of the ‘$y$ scaling’ wave vector variable, $y = (\omega - \omega_R)/v_R$ where $v_R = \hbar Q/m$, and to present the neutron inelastic data as $J(Q, y) = v_R S(Q, \omega)$. For finite $Q$, as investigated in our experiments, the struck atom does not recoil freely. Hence, corrections to the IA, generally referred to final state effects, have to be made. Including these effects as a convolution, the observed scattering function $J(Q, y)$ is,

$$J(Q, y) = \int dy' R(Q, y - y') J_{IA}(y')$$  \hspace{1cm} (2)

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

$$J_{IA}(y) = \int dk n(k) \delta(kQ - y) = n_Q(y)$$  \hspace{1cm} (3)

is the IA to $J(Q, y)$. Specifically, $J_{IA}(y)$ is $n(k)$ projected along $Q$ denoted the longitudinal momentum distribution [27].

### 3 Experimental Results

Two sets of measurements [28, 29] were performed on the MARI time of flight (TOF) spectrometer at the ISIS Facility, Rutherford Appleton Laboratory, United Kingdom. As stated above, the goal was to measure BEC in solid $^4$He. In the first experiment, data were collected on a solid $^4$He sample contained in an 18 cc cylindrical Al sample cell. The second experiment was motivated by the work or Rittner and Reppy [3]. The solid was introduced in a larger Al cylindrical cell (100 cc) but confined within a stack of Al disks with a surface to volume ratio of 40 cm$^{-1}$. Data were collected as
Fig. 1 (Color online) Observed $J(Q, y)$ at $Q = 28.0 \, \text{Å}^{-1}$ versus $y$ at temperature $65 \, \text{mK}$ (open circles). The solid line is a fit of the model OBDM. The dashed line is the MARI instrument resolution.

![Graph](image)

a function of temperature, from 500 mK down to 65 mK. We used commercial grade purity $^4$He (0.3 ppm $^3$He) and the solid was grown from liquid in the cell using the blocked capillary method. An incident neutron energy of 750 meV was used and data were collected at constant scattering angles. The data were converted to the dynamic structure factor $J(Q, y)$ at constant wave vector transfer $Q$ in the range $20 \leq Q \leq 29 \, \text{Å}^{-1}$ and energy transfer ($\hbar \omega$) expressed in the $y$ variable, described above.

Figure 1 shows the observed $J(Q, y)$ at wavevector $Q = 28 \, \text{Å}^{-1}$ and temperature $T = 65 \, \text{mK}$. The observed $J(Q, y)$ includes the simulated MARI instrument resolution function which is shown separately as a dotted line in Fig. 1. The excellent resolution on MARI is highly desirable, and allows an accurate determination of the $^4$He kinetic energy, which is set by the width of $J(Q, y)$. The solid line is a fit of a model to the data as described below.

To obtain a condensate fraction, we assumed a model $n(k)$ in (3) of the form,

$$n(k) = n_0 \delta(k) + (1 - n_0) n^*(k),$$

(4)

where $n^*(k)$ is the momentum distribution of the atoms above the condensate in the $k > 0$ states. To proceed, we assume (1) that the shape of $n^*(k)$ is the same below and above $T_c$ and (2) that the FS function $R(Q, y)$ in (2) is the same as observed previously [30] in liquid helium. The free parameters (two) in the model are then $n_0$ and the width, $\bar{\alpha}_2$, of the Gaussian component of $n^*(k)$. A condensate component appears as an additional intensity $n_0 R(Q, y)$ in $J(Q, y)$ [30].

Figure 2 shows our observed condensate fraction $n_0$ as a function of temperature for the two measurements. In the first measurement, we found a condensate parameter which is zero within 1% at 80 mK. With an improved precision and larger sample in the second experiment, we found a condensate parameter which is again zero within 0.3%. This negative result suggests that either $n_0$ is small even where $\rho_S/\rho$ is significant or the large $\rho_S/\rho$ may be associated with rapid initial cooling rather than large $S/V$. It also suggests that $n_0$, if present, is lower than 0.3%. This result is our key finding.
Fig. 2  Observed condensate fraction $n_0$ (in %) below $T_c$ as a function of temperature. The solid circles summarize the results of our first measurement on a bulk solid $^4$He sample in an 18 cc Al sample cell. The open circles show the results of our second measurement, where the solid was grown in a larger Al sample cell (100 cc) having a surface to volume ratio of 40 cm$^{-1}$.

4 Conclusions

In summary, we have not observed a non-zero condensate fraction, within the precision of our neutron scattering data, and can not confirm at this time that NCRI is associated with BEC. As a reference, we note that measurements $^{30}$ on MARI provide the current best value of the BEC condensate fraction $n_0$ in bulk liquid $^4$He at zero pressure. Future searches for BEC in solid $^4$He will thus require higher precision, and lower temperatures. Studies of the effects of sample quenching on the NCRI would be particularly useful. We expect to gain better statistics on $n_0$ using a higher flux neutron spectrometer, such as the ARCS instrument at the Spallation Neutron Source (SNS). The flux at ARCS is estimated to be an order of magnitude greater than that at MARI. If a non-zero condensate fraction is observed in a solid, this would be an exciting discovery in condensed matter physics. If on the other hand, we can improve our measurements to reach a precision on $n_0$ of ±0.05% and BEC is still not observed, then we can begin to eliminate amorphous regions and high concentration of vacancies as a possible mechanism of superflow in solid $^4$He. In any case, future neutron scattering experiments will provide decisive information on this fascinating new phase.

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References