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Neutron scattering study of the excitation spectrum of solid helium at ultra-low temperatures

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Abstract. There has been a resurgence of interest in the properties of solid helium due to the recent discovery of non-classical rotational inertia (NCRI) in solid ^{4}He by Chan and coworkers below 200 mK which they have interpreted as a transition to a 'supersolid' phase. We have carried out a series of elastic and inelastic neutron scattering measurements on single crystals of hcp ^{4}He at temperatures down to 60 mK. While we have found no direct evidence of any change in the excitation spectrum at low temperatures, we have found that the excitation spectrum of solid ^{4}He shows several interesting features, including extra branches in addition to the phonon branches. We interpret these extra branches as single particle excitations due to propagating vacancy waves, which map on to the famous 'roton minimum' long known in the excitation spectrum of superfluid liquid ^{4}He. The results show that in fact solid ^{4}He shares several features in common with the superfluid.

Keywords. Solid helium; phonons; vacancy excitations; roton.

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1. Introduction

Recently, there has been renewed interest in the possibility of the existence of a 'supersolid' He phase. Theoretical speculation about possible Bose–Einstein condensation (of zero-point vacancy excitations) and superfluidity in solid ^{4}He began almost 40 years ago [1–4]. Early experimental searches, using macroscopic techniques, found no evidence for the 'supersolid' state [5,6]. However, evidence for some type of phase transition in impure or strained crystals was found using ultrasonic propagation [7,8]. Recent observations of a decrease in the moment of inertia of the solid, in impure or strained crystals, at temperatures below about 200 mK were interpreted as evidence for a supersolid [9–11]. However, an attempt to force direct flow through small channels was unsuccessful [12], and deep inelastic neutron scattering finds no evidence for a zero-momentum condensate fraction [13,14].

As in the case of superfluid helium, the excitations in the solid could provide microscopic evidence for a possible Bose condensate or supersolid behaviour if present. There is no direct experimental evidence for the existence of zero-point vacancies, and quantum Monte-Carlo calculations of perfect crystals do not find such vacancies [15,16].

We report here inelastic neutron scattering measurements on low density solid ^{4}He crystals in the hexagonal close packed (hcp) phase. The measurements reveal portions of the spectrum that were not previously observable, were done at lower temperatures and provide higher resolution than in earlier experiments [17,18]. We find no evidence for a phase transition within the solid state but we do provide evidence for dispersive non-phonon excitations. We identify these excitations as waves of delocalized vacancies. Certain similarities between parts of the vacancy wave excitations and the roton spectrum in the superfluid are suggestive of strong links between the solid and liquid phases.

2. Experimental results

Measurements of the inelastic spectrum were made on the NG4 Disk Chopper time-of-flight spectrometer at the NIST Center for Neutron Research in Gaithersburg, MD. Experimental details will be published elsewhere. Measurements were made both for the liquid in the superfluid phase as a function of Q and for the solid hcp phase at various temperatures below the melting temperature. No detectable change in the excitation spectrum of the solid was seen as the temperature was taken well below 200 mK, which is roughly where the so-called 'non-classical rotational moment of inertia' appeared in the torsional oscillator experiments [9].

Figures 1a and 1b show scattering intensity contours in energy–wavevector space for reduced wavevectors in the basal plane which were longitudinal and transverse respectively relative to the (1 0 0) Bragg peak for Crystal 1. Measurements were made at a few different incident neutron energies. For each incident energy, at least 21 different angles of the crystal relative to the incident beam were used, covering at least 42°. For comparison figure 1c shows the corresponding inelastic spectra for the superfluid at a pressure just below the freezing pressure (24 bar).

The dominant new features in the longitudinal cut (figure 1a) are:

- a quadratic dispersion curve centred at the (1 0 0) reciprocal lattice point.
- the apparent intersection and subsequent disappearance of the two sharp longitudinal modes about half way to the zone boundary. This is shown in more detail in figure 1d and discussed below.
- a marked asymmetry in the intensity of the acoustic phonon with respect to $+q$ and $-q$ (where q is the reduced wavevector measured from the (1 0 0) Bragg point). This has been observed in previous studies of solid ^{4}He [17,18].
- a broad spectral feature that appears at about 2 meV, for wavevectors beyond the point of intersection mentioned above, and extends beyond the energy and wavevector range measured here. This latter broad feature is also seen in the superfluid (figure 1c), starting at a similar absolute momentum transfer ($\approx$2.4 Å^{-1}) and a slightly lower energy, corresponding to approximately twice the

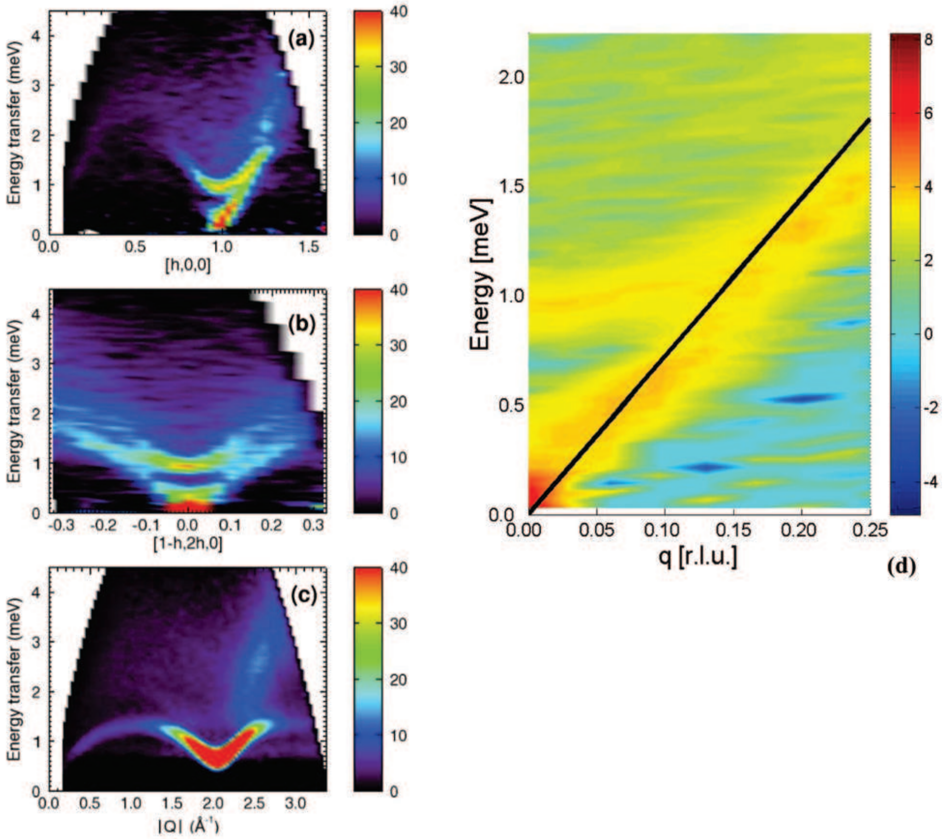

Figure 1. Cuts of scattered intensity contours in energy–wavevector space for hcp ^{4}He. (**a**) For Q along $\langle 1\,0\,0\rangle$, (**b**) for Q through the $(1\,0\,0)$ reciprocal lattice point in the transverse direction in the basal plane, (**c**) for the superfluid at $P = 24$ bar and $T = 0.22\,$K. Intensities have been integrated over the perpendicular direction to the cut by 0.01 reciprocal lattice units. (**d**) Expanded plot of cut shown in (a) to show detail of 'collision' and merging of upper longitudinal mode and LA phonon mode.

minimum energy gap in each case. This feature is also seen in the normal fluid [19]. In the solid, the high energy excitation displays hexagonal symmetry about the origin. In ref. [19], this scattering was identified as broad, multiphonon scattering. Alternatively, it might also be associated with strongly interacting single-particle scattering, in which case it should connect with the free particle recoil scattering at larger momentum transfers. Comparing figures 1a and 1c, the spectra appear superficially to be very similar.

The longitudinal data were all confirmed on Crystal 2. We now examine the low energy excitations close to the $(1\,0\,0)$ Bragg point more carefully. Figure 2 shows the dispersions of these excitations, as obtained by fitting Gaussians to the data. The lowest energy mode is an acoustic phonon and we argue that the sharp intermediate

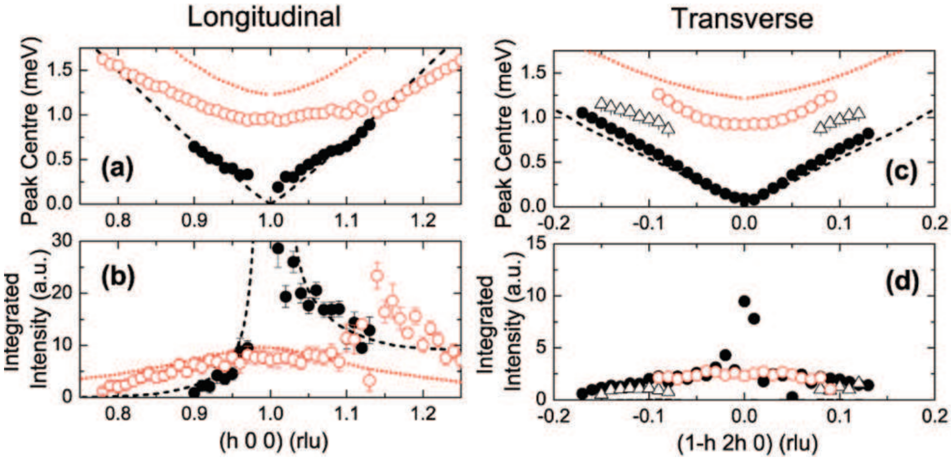

Figure 2. The sharp excitations in the longitudinal and transverse directions from the (1 0 0) Bragg peak, measured at 0.055 K for the longitudinal cuts (**a,b**) and for the transverse cuts (**c,d**). The positions (a,c) and intensities (b,d) were obtained from Gaussian fitting. The acoustic phonon branches are shown by filled circles, the vacancy mode by open circles and in the transverse case there is an additional mode marked by open triangles. In the upper panels (a,c), the lines are the calculated dispersion curves for the acoustic (dashed) and optic (dotted) phonon branches from the self-consistent phonon calculations in ref. [20]. In (b), the intensities are calculated from the inelastic structure factor following ref. [20] with an additional common scaling factor.

energy mode is a propagating vacancy excitation. Minkiewicz *et al* [17] associated the intermediate energy mode with an optical phonon despite the discrepancy with the self-consistent phonon calculations of Gillis *et al* [20], to be discussed below.

The intensities for the longitudinal branches were calculated from the inelastic structure factor based on this self-consistent phonon theory [18,20], and compare favourably to the data in figure 2 except at the intersection. In particular, the asymmetry in the intensity of the acoustic phonon branch phonons is well reproduced.

We present two arguments for associating the intermediate energy mode with a vacancy excitation rather than an optical phonon mode:

(i) This mode intersects with the longitudinal acoustic phonon branch. On symmetry grounds, longitudinal optic and acoustic phonon branches in an hcp structure may not cross or degenerate except at the zone boundary in the c direction. For the transverse modes, where the zone boundary occurs at $h = 0.33$, there is no collision between the strongest modes, presumably due to the lower transverse sound velocity.

(ii) If the positions of the optic and acoustic branches are taken from the self-consistent phonon calculations and the existence of a third dispersive excitation is assumed, excellent fits to the data can be obtained using damped harmonic oscillator line shapes convoluted with a Gaussian instrumental resolution function of 0.2 meV FWHM. The actual optic branch corresponds

> to the broad, weak excitation at higher energies, with a significantly broadened linewidth. This assignment resolves the discrepancy between the self-consistent phonon calculations and experimental observations. Close to their intersection, both the acoustic branch and the vacancy mode widen significantly, indicating decay of phonons into vacancy waves. Above the intersection, just one sharp mode persists. Markovich *et al* [21] observed an optic-like mode in the bcc phase of ^{4}He in a longitudinal cut through the (1 1 0) Bragg peak, although none should exist in a monatomic bcc structure. The strong similarity between this spectrum and that reported here in the hcp phase suggests that this branch of the spectrum in the bcc phase could be the vacancy excitation.

With our identification of the intermediate energy mode as a vacancy excitation, the minimum energy is identified as the vacancy activation energy, and is found to be 11 K. This is consistent with values obtained by NMR [22] and observation of activated thermal expansion above ≈10 K in X-ray [23] and neutron diffraction [24] studies. The higher activation energy observed in the bcc phase [22] is consistent with the higher melting temperature of the bcc crystal. The dispersion of this vacancy mode is quadratic, with an effective mass of $\sim 0.11 m_{\mathrm{He}}$. This excitation was resolution limited, but from our highest resolution data with incident neutron wavelength 5.2 Å the maximum vacancy excitation linewidth is 21 μeV. The possibility of a dispersive mode associated with defects (specifically ^{3}He impurities) in ^{4}He has been discussed by Manousakis [25], although we note that the amount of ^{3}He in our sample is small.

To summarize, we assert that this excitation is a delocalized vacancy mode because the longitudinal optic and acoustic modes cannot cross and annihilate on symmetry grounds. In addition, a similar mode was observed in the bcc phase [21], which cannot possess an optic mode, and the dispersion of the optic mode as calculated from self-consistent phonon theory [20] is inconsistent. In fact, a broad dispersive excitation that more closely matches the calculated dispersion has been observed.

An additional mode is observed between the transverse acoustic and vacancy modes (figure 1b). This new mode possesses the lattice symmetry and therefore cannot belong to a second crystal, a possibility also dismissed by Laue scans of the crystal. This dispersive mode is seen at two different incident wavelengths, 5.2 Å and 3.7 Å, indicating that it is not due to multiple scattering. The origin of this mode is currently not known.

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