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Temperature dependence of the collective excitations of liquid ^{4}He at high pressure

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Abstract. Systematic high-resolution neutron inelastic scattering measurements have been made of the collective phonon–roton excitations in normal and superfluid ^{4}He as a function of both pressure (density) and temperature. The IN6 time-of-flight spectrometer at the Institut Laue–Langevin has been used to obtain accurate and continuous data over a wavevector range 0.3 Å^{-1} $< Q <$ 2.35 Å^{-1} with a resolution of 0.12 meV. We present the experimental data and discuss the energies and widths of the single excitations; changes concomitant with the λ transition are discussed. The structure of the multiphonon continuum at pressure is also discussed and is shown to be consistent with theory.

1. Introduction

The superfluid properties of liquid ^{4}He below the λ transition (2.17 K at saturated vapour pressure (SVP); 1.93 K at 20 bar pressure) lead to the existence of sharp, well defined collective excitations with a 'roton' minimum [1]. Neutron inelastic scattering allows direct observation of these excitations. We refer the reader to the monographs by Glyde [2] and Griffin [3] for overviews of the many neutron scattering studies of superfluid ^{4}He to date. The IN6 spectrometer at the Institut Laue–Langevin has been used to perform high-resolution neutron inelastic scattering measurements of the pressure and temperature dependences of the collective excitations in ^{4}He. In conjunction with the temperature-dependent study at SVP by Andersen *et al* [4], S(Q,E) has now been obtained over a major part of the normal and superfluid phase diagram under nearly identical resolution conditions. The measurements presented here focus on the superfluid phase. They therefore complement the measurements of Crevecoeur [5], which focus on the normal fluid at pressure.

In this paper we present a preliminary report of the temperature-dependent data from liquid ^{4}He at 20 bar pressure. These data are typical of the temperature dependence of $S(Q,E)$ near the solidification pressure (25 bar at 0 K). Attention is focused on the phonon, maxon and roton regions. A qualitative discussion of these is given. In addition the Q and E dependences of the multiphonon excitations are also discussed.

The effect of pressure on the excitations at low temperatures has been detailed in previous studies [6–8]. In the phonon region the gradient $(\mathrm{d}E/\mathrm{d}Q)$ of the dispersion is

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larger at high pressures; in accordance with the increase on the first sound velocity c. The excitation energies increase for increasing wavevector up to nearly the roton minimum. Thereafter they decrease. The wavevector of the roton minimum is larger at high pressures. Previous high-resolution studies of the temperature dependence of the collective excitations [6, 7] led to a new interpretation by Glyde [2] and Griffin [3] showing a link to Bose–Einstein condensation. In the present study, quantitative changes were shown to occur at the λ transition at all pressures and this implies that the excitations are indeed related to the existence of a condensate.

2. Experimental details

An incident neutron energy of 4.75 meV was chosen on the IN6 time-of-flight spectrometer. An extensive detector bank of over 300 detectors covering scattering angles from 10° to 115° allowed continuous coverage of wavevectors between 0.3 $\text{Å}^{-1} < Q < 2.35$ Å^{-1}. Typical inelastic resolution widths were about 0.15 meV (full width at half-maximum). The high-purity ^{4}He sample was held in a cylindrical aluminium sample cell of 24 mm diameter, with vertically spaced cadmium discs placed inside at 10 mm intervals to minimize multiple scattering. The following pressures were measured at 500 mK at SVP: 2, 5, 10, 15 and 20 bar (a measurement was also performed at 30 bar in the solid phase). An extensive study of the temperature dependence covering 12 temperatures below and above T_λ was made at 5, 10, and 20 bar pressure. The scattering from the empty cell at low temperatures less than 4 K was measured. The incoherent scattering from vanadium was also measured to correct for the various detector efficiencies. The vanadium and the SVP ^{4}He data were also used to calibrate the spectrometer's incident neutron energy and sample to detector distances. As the data were recorded at a constant scattering angle, an algorithm was devised [4] to re-bin the data onto strips of constant Q for easier visualization and comparison with theory.

3. Temperature dependence of $S(Q, E)$ at 20 bar

$S(Q, E)$ for the phonon, maxon and roton at 20 bar are shown in figures 1, 2, and 3 as a function of temperature. The temperature dependence of the phonon ($Q = 0.4$ Å^{-1} (figure 1)) at high pressures is, in many respects, similar to the temperature dependence of the phonon at SVP. The low-temperature scattering consists of a single sharp peak with very little observable multiphonon scattering. Below T_λ, the single-phonon peak position remains constant and broadens increasingly with increasing temperature. However, in contrast with the SVP data, there is a more marked increase in the phonon energy (frequency) at the λ transition. The phonon excitation changes little above T_λ.

The change in the phonon energy at T_λ when the helium is at a high pressure is significant in view of the Glyde–Griffin model [2, 3]. Within this model the zero-sound-like regular density model (not believed to undergo dramatic changes at the (λ transition) is coupled with a quasi-particle mode proportional to n_0 (the Bose-condensate fraction) and is not therefore present above T_λ. The conventional interpretation of this model is that at higher pressures these models become decoupled and are distinctly separable and divergent as $Q \to 0$ [3]. The increased change in the single-phonon energy at $Q = 0.4$ Å^{-1} at T_λ challenges this conventional interpretation. The more marked changes to the phonon at T_λ indicate that a contribution from the quasi-particle mode must still be present in the phonon region. Moreover, this coupling from the quasi-particle mode into the single phonon must *increase* with increasing pressure.

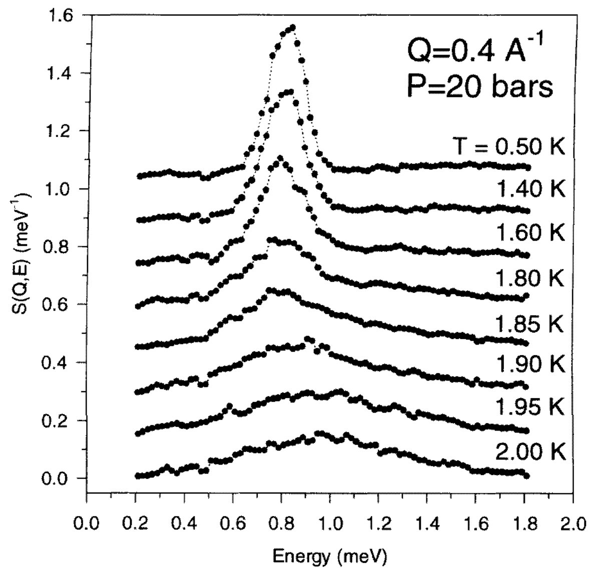

Figure 1. $S(Q, E)$ at $Q = 0.4$ Å^{-1}, $P = 20$ bar showing the temperature dependence below and above T_λ.

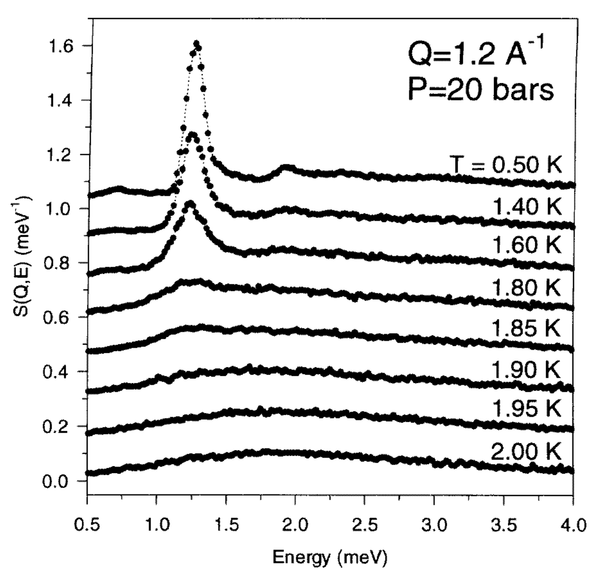

Figure 2. $S(Q, E)$ at $Q = 1.2$ Å^{-1}, $P = 20$ bar showing the temperature dependence below and above T_λ.

At the maxon wavevector ($Q = 1.2$ Å^{-1}), the low-temperature single-phonon excitation is superimposed on a (largely temperature independent) multiphonon continuum. The magnitudes of multiphonon excitations are significant compared with the single-phonon excitation peak. The low-temperature single-phonon excitation peak broadens and

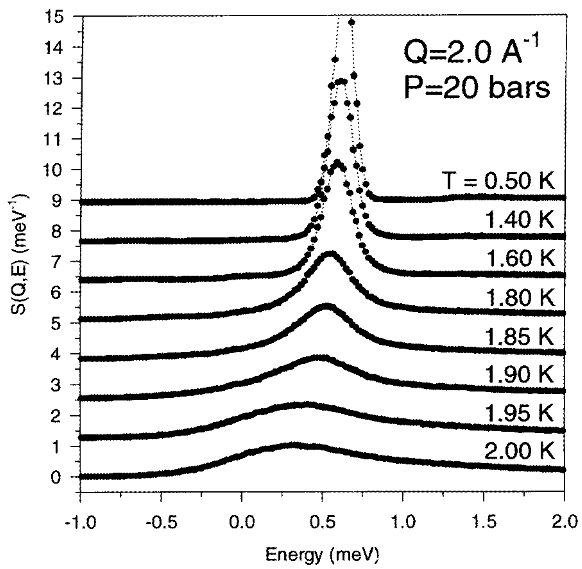

Figure 3. $S(Q, E)$ at $Q = 2.0$ Å^{-1}, $P = 20$ bar showing the temperature dependence below and above T_λ.

diminishes with increasing temperature while remaining at constant energy. Above T_λ it has disappeared entirely, leaving broad, largely temperature-independent scattering.

The intensity of the single-roton peak ($Q = 2.0$ Å^{-1}) is much greater than that of the maxon. The energy of the intense sharp single-roton excitation at low temperatures decreases with increasing temperature. The sharp peak broadens with increasing temperature, and above T_λ the broad scattering is peaked at a lower energy than the single sharp peak present in the superfluid. Although multiphonon scattering is observed in the roton region, the single-phonon excitation scattering is dominant. At all Q, the high-energy tail to $S(Q, E)$ is independent of temperature.

4. Structure of the multiphonon continuum at high pressures

Figure 4 shows a contour map of the multiphonon excitations at $T = 500$ mK and $P = 20$ bar. The full height of the single excitations has been 'chopped off', revealing the Q and E dependences of the (considerable) multiphonon structure. At the applied pressures, because the maxon and the roton are further apart in energy, many of the multiphonon features are more distinguishable than in the SVP data of Andersen *et al* [4]. In the maxon region, a peak in the multiphonon excitation is seen at the single-phonon excitation energy E_{maxon} plus the roton energy Δ. At smaller Q, towards the phonon value, this broad 'peak' appears to converge towards $E \approx 2\Delta$ at $Q = 0$. Raman scattering measurements show the existence of a two-roton bound state at very low Q [9]. The multiphonon scattering observed in this study appears consistent with this interpretation, although our data do not extend to sufficiently low Q to draw any firm conclusions.

At roton wavevectors a 'ridge' of scattering is seen at approximate energies of 2Δ. At 20 bar this ridge of scattering is quite distinct from the near-recoil scattering appearing in the range 2 meV $< E <$ 3 meV. This two-roton 'ridge' coincides with the single-

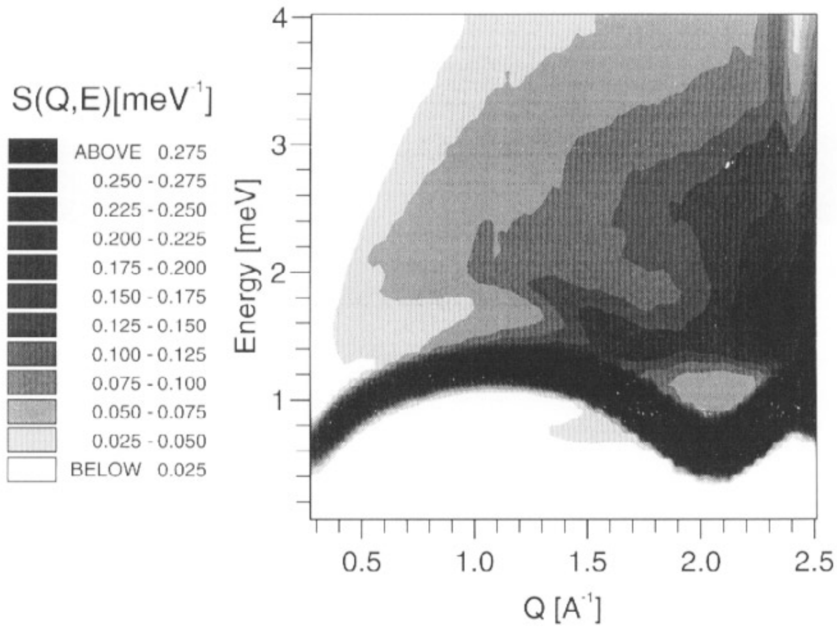

Figure 4. Contour map showing the low temperature (500 mK) multiphonon excitations at 20 bar.

phonon excitation peak at $Q \approx 1.5$ Å^{-1}, possibly leading to strong anharmonic crossover effects between single-phonon and multiphonon excitations. At maxon Q-values, this peak coincides and merges with a broader feature between the maxon+roton and the two maxon energies. The feature below the single-phonon excitations at $Q \approx 1.5$ Å^{-1}, $E = 0.75$ meV is multiple scattering. As part of their extensive study, Manousakis and Panharipande [10] used correlated basis function techniques to calculate the multiphonon structure at $T = 0$. Unfortunately their calculations have been performed only at SVP. Their calculations do show features, however, very similar to the scattering observed here and in the work by Andersen *et al* [4].

5. Conclusion

In summary, the temperature dependence of the collective excitations in ^{4}He has been shown in the phonon, maxon and roton regions. The phonon is seen to exhibit more pronounced changes at the λ transition at high pressures than at SVP. The multiphonon intensity in the maxon region is significant in comparison with the single-phonon excitation intensity. Strong coupling may exist between the multiphonon and the single-phonon excitations in this region, especially at high pressures where the maxon energy is greater than twice the roton energy, leading to possible decay processes. Although new features have been observed in the roton region, extensive high-precision data now exist on this fundamentally important region of the dispersion relation. A thorough and quantitative analysis of the data is under way.

Acknowledgments

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