Observation of a New Excitation in bcc ^{4}He by Inelastic Neutron Scattering

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We report neutron-scattering measurements of the phonons in bcc solid ^{4}He. In general, only three acoustic phonon branches should exist in a monatomic cubic crystal. In addition to these phonon branches, we found a new "opticlike" mode along the [110] direction. One possible interpretation of this new mode is in terms of localized excitations unique to a quantum solid.

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The main attribute of a quantum solid is the large zero point motion of the atoms. The vibrating atoms thus encounter the repulsive part of the interatomic potential, leading to strong short range correlations. Theoretical description of the solid is consequently a tour de force study of anharmonic effects. The self-consistent phonon (SCP) theory which emerged over the years, treats these short range correlations using Jastrow-type wave functions in the description of the ground state [1–6], in addition to the inclusion of anharmonic terms in the potential. Phonon dispersion curves calculated in the SCP theory are in good agreement with neutron-scattering experiments on solid ^{4}He [7,8], except for the $T_1[110]$ branch of bcc ^{4}He, which experimentally has a much lower energy. In addition, the SCP theory was able to explain several observations unique to quantum solids, such as multiphonon scattering which dominates the scattering already at intermediate momentum transfer Q, and phonon interference which gives rise to oscillations of the effective Debye Waller factor. Our motivation to do new experiments came following a recently proposed model [9]. This model aims to explain the discrepancy regarding the $T_1[110]$ branch of bcc ^{4}He through an approach in which the zero point motion is treated separately from the usual harmonic degrees of freedom. Indeed, the $T_1[110]$ mode predicted by the new model agrees with experiment, while leaving the other phonons unaffected. In addition, the model predicts the appearance of a new dispersionless excitation branch at an energy twice that of the $T_1[110]$ phonon at the zone boundary ($\sim$1.2 meV). Such an "opticlike" mode was not reported [7,8].

In order to check this prediction, we performed inelastic neutron-scattering experiments on the bcc phase of ^{4}He using the IN-12 spectrometer at the ILL. ^{4}He single crystals were grown and oriented in the beam. Since the maximal temperature width of the bcc phase is less than 50 mK, particular care was devoted to temperature control, resulting in a $\pm$0.1 mK stability. In addition, the growth cell contained a capacitive pressure gauge allowing us to precisely control the location on the phase diagram. Out of the five crystals grown, four could be oriented in the $\{11\bar{1}\}$ scattering plane, and one in the $\{100\}$. From the observed Bragg peaks, the cell contained only one large single crystal. The energy of the incident beam could be varied in the range of 2.3 to 14 meV. Constant-Q scans were usually done at fixed k_F. In the high resolution scans we used a cooled Be filter to remove $\lambda/2$ contamination. The highest instrumental resolution used was 0.07 meV (FWHM) with $k_F = 1.15$ 1/Å. Data were typically taken around the $(1, 1, 0)$ point.

In addition to well defined single phonon groups and broad multiphonon features, the measured spectra also show a new, well defined feature. In Fig. 1, we show an intensity map displaying longitudinal scans in which the new feature appears as a weakly dispersive opticlike branch, together with the $L[110]$ phonons. The resolution in Fig. 1 is not sufficient to display details of the spectra, which are seen more clearly in Fig. 2. The new feature was

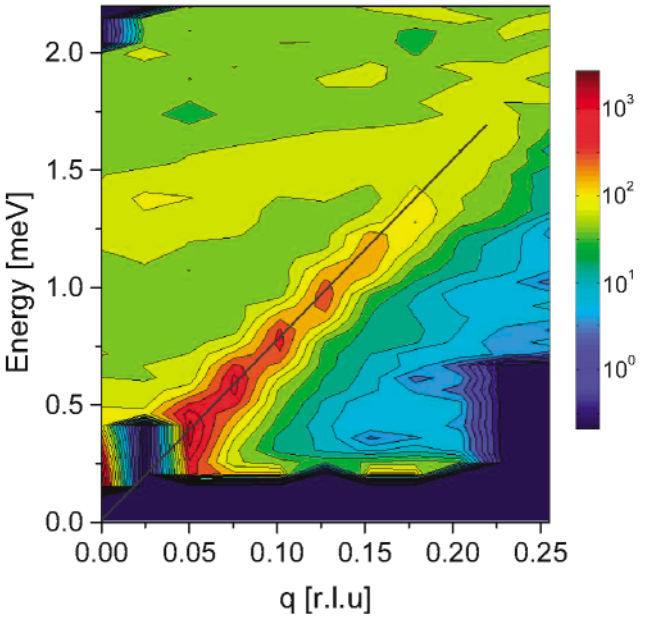

FIG. 1 (color). Intensity map of longitudinal scans along [110] showing the L branch and an "opticlike" excitation band. Note that the intensity scale in the figure is logarithmic, given in neutron counts.

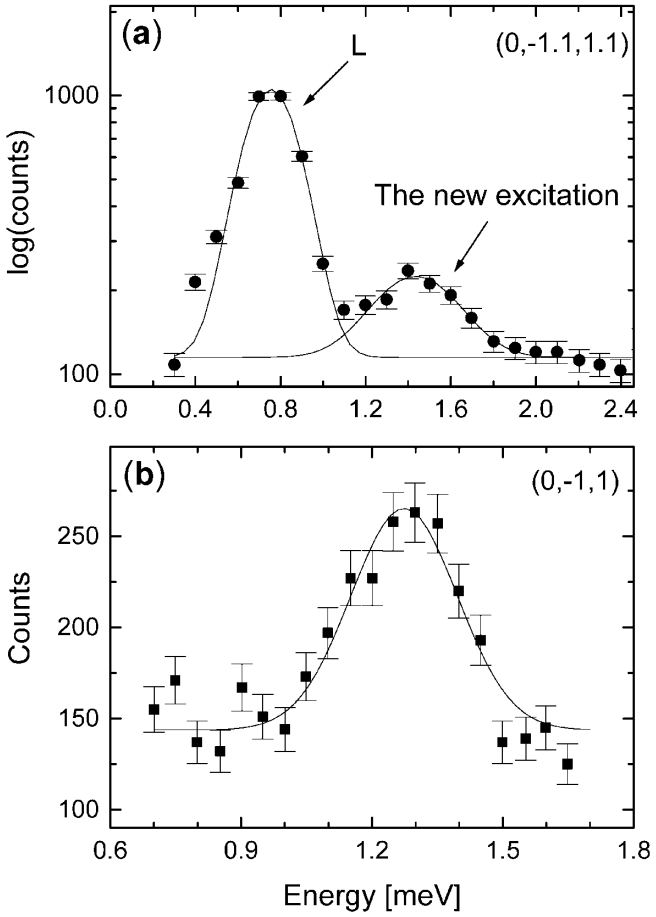

FIG. 2. Typical neutron scattering spectra along [110] showing the new feature for two values of q. In (a), taken at $q = 0.1$, the feature appears together with the L phonon. In (b), taken at the zone origin ($q = 0$), only the new feature is seen. The lines in the figure are Gaussian, and serve as a guide to the eye.

observed in all four crystals during scans along the [110] direction. This feature disappeared once the crystal was molten and the cell contained only liquid. It is therefore not connected with scattering from the liquid or from the walls of the cell. Moreover, we performed intentional experiments in the mixed phase region, with both hcp and bcc crystals in the cell. We observed no correlation between the phonon peaks of the hcp phase and the new feature. We conclude therefore that the new feature is a property of the bcc solid.

The average energy (over all four crystals) of this new mode at the zone origin is 1.23 meV, almost exactly twice the energy of the zone boundary T_1 phonon (0.59 ± 0.01 meV). This ratio is constant over the whole temperature range where the bcc phase exists. The best location to observe this mode was at a reduced wave vector $q = 0$, where it was not masked by any of the other phonon branches. However, as Fig. 1 shows, it is possible to resolve this mode up to $q = 0.2$ in units of $2\pi/a$ (r.l.u.). At higher q, the phonon groups are too broad to identify this feature. In Fig. 3, we show the dispersion relations measured along the [110] direction. The lines in the figure are from the SCP theory [10] and from the new model [9]. Our data for the three phonon branches are in excellent agreement with previous experiments [7,8]. The fact that the T_1 branch is lower by a factor of ~ 2 compared with the SCP calculation was already evident from the previous experiments [7,8], and is confirmed in the present work. In addition, Fig. 3 shows the data for the new feature, which shows some dispersion. It is important to ask whether the new feature is a distinct mode, rather than a product of anharmonic effects described by the SCP theory [11–15], namely phonon interference and multiphonon scattering. First, phonon interference gives rise to scattering intensity which is an odd function of q; namely, it vanishes at the zone origin [11]. The fact that we see this feature clearly at the zone origin excludes the possibility that it is due to interference effects. Regarding multiphonon effects, one could generate a feature at $q = 0$ and energy of 1.2 meV as a bound state of two zone boundary T_1 phonons with opposite momenta. However, in the same way one could

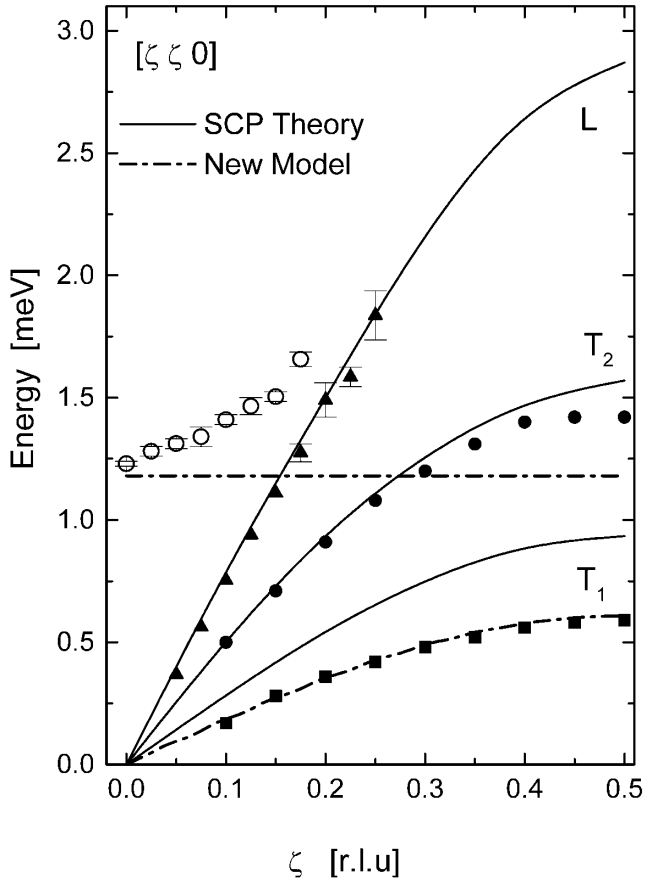

FIG. 3. Dispersion curves along [110]. Solid symbols—data for the three phonon branches. Open symbols—the new feature. Solid lines represent the SCP theory [10]. The additional predictions of the new model are shown as dash-dotted line (the prediction of the new model for the L and T_2 branches are the same as of the SCP theory).

combine different phonons to obtain any energy and q; namely, this process will produce a broad background, in contrast to what is seen in Fig. 2. Typically, the width of multiphonon features is on the order of $\sim$ several meV, while that of the new feature is an order of magnitude smaller (about 0.2 meV). Next, in Fig. 4 we plot the measured linewidth of all the phonons along (110) vs their energy. To obtain the data in this figure, we used well defined, symmetric phonon groups. The phonon groups were fit to a damped harmonic oscillator function folded with the Gaussian spectrometer resolution function determined by scattering from the vanadium sample. One can see that phonons with energy less than 1.2 meV have widths which are below our resolution limit ($\leq$0.02 meV), while above that energy there is a marked increase of the linewidth. In contrast, the linewidth predicted by the SCP theory [16] due to phonon interaction is a smoothly increasing function of energy (see Fig. 4). The abrupt increase shown by the data strongly suggests that the feature at 1.23 meV is a distinct excitation which the high energy phonons can decay into.

We outline here possible interpretation of the data involving localized excitations. Here, localized excitations can be point defects or "local modes" [17]. This approach is appealing since point defects in solid He are predicted to add another excitation branch to the usual phonon spectrum [18–20]. Interpretation of the new feature in these terms is supported by thermally activated mass [21] and spin diffusion [22] experiments in bcc ^{4}He, in which the activation energy of point defect was found to be 1.25 meV. One possibility is that the new observed mode is connected to a point defect such as vacancy, interstitial, or a Frenkel pair, involving a *displacement* of atoms from their lattice sites. Several authors proposed that point defects in quantum crystals are delocalized

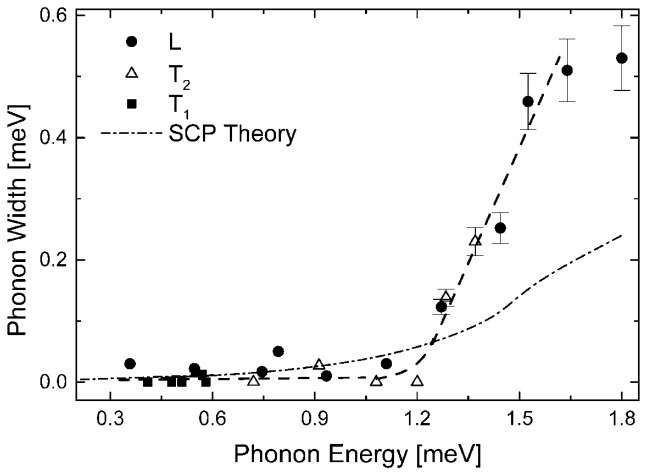

FIG. 4. Experimental phonon linewidth vs phonon energy for all three branches along [110] showing the abrupt increase near 1.2 meV. Symbols are experimental data. Dash-dotted line — SCP theory for the L phonon (Ref. [16]). The dashed line is a guide to the eye.

objects and can behave as quasiparticles, having a finite bandwidth and well defined dispersion relation [18–20]. Indeed, the data for the new feature in Fig. 2 can be fit by an expression of the type $E(q) = \epsilon_0 + \hbar^2 q^2/2m^*$, with $\epsilon_0 = 1.23$ meV and $m^* \simeq 0.2m_4$. We cannot determine the bandwidth directly because the new mode cannot be resolved for $q \geq 0.2$ r.l.u. There are, however, problems associated with this interpretation: First, according to the data shown in Fig. 4, phonons can decay into this mode once their energy exceeds ϵ_0. For the L branch, the phonon with the lowest energy ($= \epsilon_0$) that can decay has $q = 0.15$ r.l.u. If the mode has intrinsic dispersion, then there is a problem with momentum conservation. Since the decay is observed, it is possible that the new mode is intrinsically dispersionless, and the apparent dispersion seen at finite q can be attributed to mode coupling effects [23,24]. A dispersionless mode is not what these models predict [18–20]. Second, these models assume that the energy spectrum of point defects exists along every direction in the crystal. Our preliminary measurements done along the [100] and [111] directions do not show the new mode.

The other possibility is an interpretation according to the new model [9]. In this model, this new excitation represents a density fluctuation localized over two unit cells. Unlike the point defects described above, this excitation is a *dynamic* density fluctuation, in which atoms remain associated with their lattice sites. First, the model predicts correctly the energy of this new excitation at $q = 0$. Second, as Fig. 3 shows, both the L and T_2 phonons become damped above the same energy of 1.2 meV. For the L phonon this happens at $q = 0.15$ r.l.u., while for the T_2 phonon the same thing happens at $q = 0.27$ r.l.u. Hence, the new feature seems to be essentially dispersionless, as predicted [9]. However, one problem with this interpretation is that coupling of the new mode with other phonons, leading to the apparent dispersion and phonon decay, is not treated within the model.

Finally, model calculations done on strongly anharmonic solids indicate the possible existence of localized modes [17]. It would be interesting to extend these calculations using a potential more specific to solid ^{4}He, to allow a comparison with experiment.

In conclusion, we have observed a new excitation in bcc ^{4}He. One interpretation which we suggest views this excitation as a localized excitation unique to a quantum crystal. The nature of the localized excitation relies on the specific way in which atomic zero point motion is incorporated into the theoretical models [9,18–20].

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