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Opticlike excitations in bcc ^{4}He: An inelastic neutron scattering study

O. Pelleg,[1,*] J. Bossy,[2] E. Farhi,[3] M. Shay,[1] V. Sorkin,[1] and E. Polturak[1,†]
[1]*Physics Department, Technion-Israel Institute of Technology, Haifa 3200, Israel*
[2]*CNRS-CRTBT, Boîte Postale 166, 38042 Grenoble Cedex 9, France*
[3]*Institut Laue Langevin, Boîte Postale 156, 38042 Grenoble Cedex 9, France*
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We report neutron scattering measurements on body-centered-cubic solid ^{4}He. We studied the phonon branches and the recently discovered "opticlike" branch along the main crystalline directions. In addition, we report another, dispersionless opticlike branch at an energy around 1 meV (~11 K). The properties of the two opticlike branches seem different. Since one expects only three acoustic phonon branches in a monoatomic cubic crystal, these new branches must represent different types of excitations. One possible interpretation involves localized excitations unique to a quantum solid.

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Elementary excitations of quantum solids show unique properties arising from the large zero-point vibration of the atoms. The interatomic potential is highly anharmonic, and the atoms feel strong short-range correlations, due to the repulsive part of the potential.[1–3] The "self-consistent phonon" theory developed to treat this problem yielded phonon dispersion curves in a reasonable agreement with experimental data available at the time.[4,5] Recently, an additional, "opticlike" excitation branch was discovered[6] along the [110] direction of a body-centered-cubic (bcc) ^{4}He. This is puzzling, since on general grounds, only *acoustic* phonon branches should exist in a monoatomic cubic crystal. Several interpretations of the opticlike excitation were discussed[6] in terms of multiphonon effects,[3] or localized excitations unique to a quantum solid. The latter include an isotropic vacancy band[7–9] or anisotropic local modes in the [110] direction associated with a correlated zero-point motion.[10] An analysis of the data showed that multiphonon effects were not the source of this branch.[6] In addition, each of the models involving localized excitations was consistent only with some facet of the data.[6] Localized excitations in solid He are of particular interest in view of the recent reports of the "supersolid" phase.[11] Our motivation to do additional experiments was twofold: first, the opticlike branch was measured only along the [110] crystalline direction. The anisotropy of this branch can be determined only if it is investigated in all directions. Second, since this branch was largely unexpected, perhaps other such features may be found.

In that spirit, we performed inelastic neutron scattering experiments on bcc ^{4}He using the IN-12 and IN-14 triple axis spectrometers at the Institut Laue Langevin. Single crystals of high-purity ^{4}He (less than 0.1 ppm ^{3}He) were grown and oriented in the beam. The growth orientation of these crystals was *a priori* random. In most cases, the limitations of orienting the crystals by tilting the cryostat allowed us to measure only along the [110] direction. In order to access other crystallographic directions, we added a homebuilt cold goniometer to the sample stage. Two versions of the apparatus were built, the first one allowing an additional ±12° tilt of the cell in an arbitrary direction. This apparatus was used for inelastic scattering along the [110] direction carried out on the IN-14 spectrometer. The second version, allowing an additional ±45° tilt along one axis, was used for measurements along [100] and [111] done in a separate experiment on the IN-12 spectrometer. Crystals of low density ^{4}He (21 cm^3 molar volume), several cm^3 in size were grown from the superfluid at $T=1.640$ K, where the temperature width of the bcc phase is maximal (~50 mK). In order to get a high-quality single crystal, the solid was further annealed overnight on the melting curve. The final crystal was composed of two large grains misaligned by 20′, with volumes of about 2 and 4 cm^3. The full width at half maximum (FWHM) of the rocking curve was about 30′ and 40′ for the two grains. Scattering experiments were conducted at the same temperature. In addition to the crystal, the cell contained a small amount of superfluid helium (less then 1% of the volume) in order to reduce any temperature gradients across the sample. In particular, we were able to get very high-resolution data along [001] and [111] from the *same* crystal, which could be oriented to access either the {002} scattering plane or the $\{1\bar{1}2\}$. The energy of the incident beam could be varied in the range of 2.3–14 meV. Constant-Q scans were done at a fixed momentum of the scattered neutrons (k_F). In the high-resolution scans, we used a cooled Be filter to remove $\lambda/2$ contamination. Focusing techniques were used to enhance the scattering intensity. The highest instrumental resolution was 0.1 meV (FWHM) with $k_F=1.2$ Å^{-1}. In order to ascertain that the features described in the following are not spurious, we carried out extensive background measurements by repeating the inelastic scans in the various directions once with an empty cell, and once with the cell full of liquid. We found that the new features disappeared once the crystal was molten, hence they are not connected with scattering from the liquid or from the walls of the cell. Bragg scattering measurements of the crystals failed to show any traces of the hexagonal close-packed (hcp) phase. We conclude, therefore, that the features are a property of the bcc solid.

Our measurements of the phonons fill some gaps in the available data for the phonon dispersion curves.[4,5,12] These results will be described in detail elsewhere.[13] In this paper, we focus on the opticlike branches. In order to assign the various peaks seen in the neutron scattering data, we per-

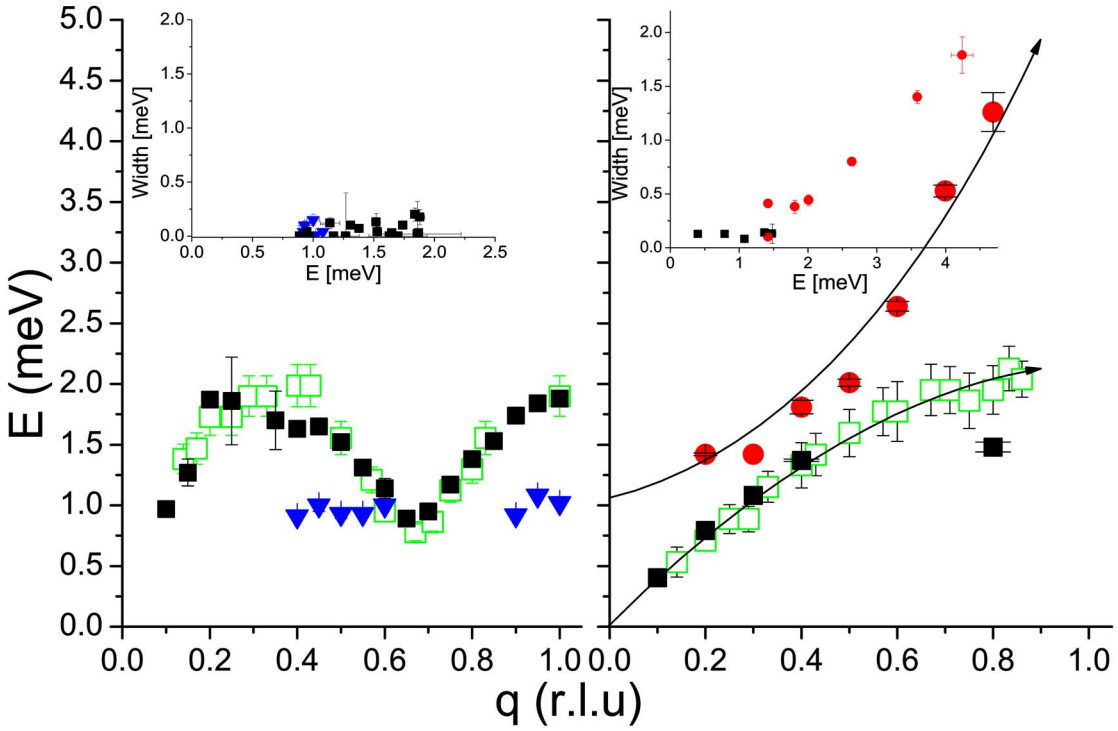

FIG. 1. (Color online) Dispersions curves along the [111] direction. Left panel: longitudinal polarization. Right panel: transverse polarization. In both panels, solid black squares are data points for the phonon branches, and the open green squares are results of the PIMC simulations of the dispersion curves (Ref. 14). The inverted blue triangles in the left panel are data points for the LOB. In the right panel, the red circles are data points for the HOB. The solid lines are a guide for the eye. The inset shows the energy linewidth of the features seen in the inelastic spectra. Note that broadening occurs only for the polarization where the HOB is observed (right panel).

formed path integral Monte Carlo (PIMC) simulations[14] of bcc ^{4}He to determine the contribution of single phonons to the dynamic structure factor $S(q,\omega)$. Figure 1 shows the dispersion relations of the L and T phonons along [111], along with the two opticlike branches observed in these experiments. The left panel shows the L branch and another, dispersionless branch at an energy around 1 meV (~11 K). We label this branch as the "lower opticlike branch" (LOB). Regarding the phonons, the data for the L branch is in excellent agreement with the simulations. The measured energy linewidth of the phonon peaks in the spectra (see inset) is small, limited by the experimental resolution. Turing to the right panel of Fig. 1, again two branches are seen, the T[111] phonon branch and another, opticlike branch, which we identify with the branch observed previously[6] along [110]. We label this branch as the "higher opticlike branch" (HOB). It is seen that the data for the T branch agree with the simulations only for energies less than the minimum energy of the HOB branch. Similarly, the inset shows that the linewidth increases quite strongly above this minimum energy. Quite obviously, one cannot assign the points on the dispersion curve with a large linewidth to single phonons. The increase of the linewidth was also observed for the L[100] branch where the HOB excitation was also seen.

First, we discuss the HOB opticlike excitation branch, previously observed only along the [110] direction.[6] In the present work, this branch was observed in scans measuring the $T(111)$ and $L(100)$ phonons, while in the scans for $L(111)$ and $T(100)$, it was absent. Hence, this excitation is anisotropic. The dispersion of this branch, when plotted together with the usual phonons, is suggestive of mode coupling. This is evident, for example, in the right panel of Fig. 1 showing the T[111] branch, as well as in earlier data[6] along [110]. Additional support for this suggestion comes from the results plotted in Fig. 2 showing the polarization dependence of the HOB along [110]. It is seen that in scans with the $T1$ polarization, this branch (labeled HOB-$T1$) shows little dis-

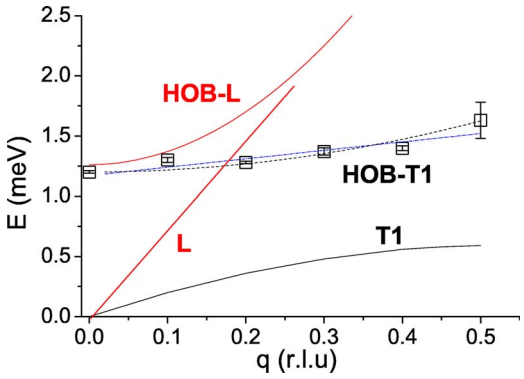

FIG. 2. (Color online) Dispersion curves of the HOB along [110] with two polarizations. The lines labeled HOB-L and L are smoothed values of the HOB and the $L(110)$ phonon branches from Ref. 6. HOB-$T1$ denotes data points from this work for the HOB with $T1$ polarization, while the line labeled $T1$ represents smoothed values of the $T1(110)$ phonon branch. The dashed black line is a linear fit to the HOB-$T1$ data, and the dashed-dotted blue line is a quadratic fit.

persion, while in scans with a longitudinal polarization (labeled HOB-L) the dispersion is significant. Since the maximal energy of the $T1[110]$ branch is 0.6 meV (~7 K), while the minimal energy of the HOB is 1.23 meV (~14 K), the $T1$ and HOB branches do not cross. Hence, mode coupling with the $T1$ branch should be weak. On the other hand, the HOB and the $L(110)$ branch do cross and so the coupling with the $L(110)$ should be stronger. Both the broadening shown in the right panel of Fig. 1 and the data in Fig. 2 are consistent with this idea.

In order to determine the intrinsic dispersion of this branch, we attempted to simultaneously fit the dispersion relations of the HOB excitation and the phonon branches using a mode coupling approach. We took the coupling of the branches to be in the form $\Delta/[(\omega_{\mathrm{HOB}}^2-\omega_{ph}^2)+iD]$, where Δ is the coupling constant, ω_{HOB} and ω_{ph} denote the energies of the HOB and phonon branches, respectively, and D is connected to the damping, which prevents a singularity at the mode crossing. This particular form is not based on a specific model of this excitation, hence it is useful only to gauge the relative strength of the coupling between the various branches. After trying several similar forms,[15,16] two conclusions can be drawn; first, it was possible to fit the coupled branches only by assuming that the HOB has a finite dispersion. Second, the value of the coupling constant Δ is different for different phonon branches. For the [110] direction, it is about twice as big as for the other directions. The ratio Δ/D, however, is approximately constant for all the branches. Consequently, we believe that the intrinsic dispersion of the HOB is that observed with the $T1(110)$ polarization (Fig. 2). The dispersion is weak, and within our resolution can be fitted to either a linear or a quadratic dependence on q. The linear fit, $E(q)=\epsilon_0+v_G q$, yields a group velocity $v_G\sim 68$ m/s.The quadratic fit, $E(q)=\epsilon_0+\hbar^2q^2/2m^*$, appears marginally better, with $\epsilon_0=1.20$ meV and $m^*\simeq 0.7m_4$. The total bandwidth is about 0.4 meV (~4.6 K). To conclude, the HOB branch is anisotropic, in the sense that it is observed only in certain directions, and seems to couple more strongly to phonons along [110]. This branch has a finite intrinsic dispersion.

Now we discuss the second feature, namely, the LOB (see left panel Fig. 1). This excitation branch appears dispersionless within our resolution, with an energy of 0.95 ± 0.1 meV [~11 K]. Typical scans showing the LOB are plotted in Fig. 3. This excitation was seen in scans measuring the $T(100)$, $L(100)$, and the $L[111]$ phonons, while being absent in scans along the [110] direction, even at the highest resolution.[6] Hence, it is also anisotropic in the same sense as the HOB branch. In this context, we also tried to examine the possibility that this mode is excited only when the incident beam is along some specific direction. We found that in the {002} scattering plane, the excitation was observed when the incident beam was around the (110) direction, while in the $\{1\bar{1}2\}$ plane the incident beam was around the (021) direction. These directions are not equivalent, so the excitation of the mode does not seem to be linked to the direction of the incident neutrons. Another remark is that near the minimum of the dispersion curve of $L[111]$ ($q=2/3$ r.l.u.), there is an inherent mixing of the $L[111]$ phonons with phonons originating in the {112} scattering planes. Hence, these planes may contribute to the intensity of the $L[111]$ phonons and perhaps add another feature to the scans. However, the LOB is also seen in the [100] direction, where there is no phonon mixing. Additional discussion of these points will be presented elsewhere.[13] The energy linewidth of this new feature is small, limited by the instrumental resolution. The intensity of the LOB decreases with reduced resolution, and at low resolution it was not observed. This may perhaps explain why it was not seen in the past.[4] It may also be the reason why the LOB was not seen in the scans measuring the $T[111]$ branch, where we were not able to work with a high resolution. The fact that the LOB is dispersionless and its linewidth remains small even when it crosses a phonon branch implies that there is no interaction between the LOB and the phonons. In that respect, the LOB and the HOB are different types of excitations.

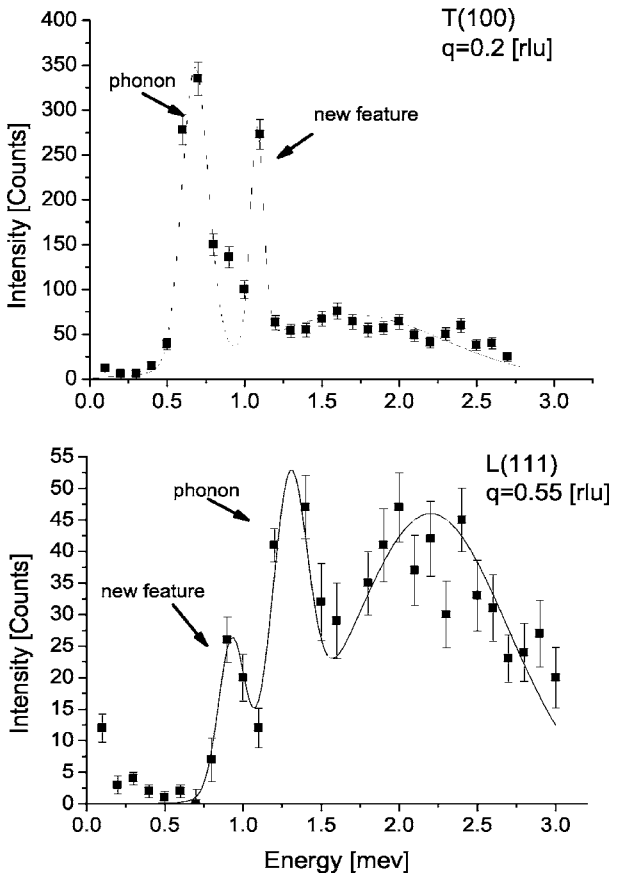

FIG. 3. Neutron groups in the [100] and [111] directions showing the LOB excitation. The squares are data points and the solid line is a Gaussian fit. The "feature" marks the LOB. The instrumental resolution in the upper panel is higher.

The absence of dispersion of the LOB suggests that this excitation is localized. Localized excitations can be point defects or more complex entities, e.g., "local modes".[17] In usual materials, phonon energies are in the meV range, while those of point defects are in the eV range. Consequently, in usual materials, point defects cannot be observed in scans measuring phonon branches. In solid He, these two energy scales are very similar, so in principle, point defects could be

excited by cold neutrons. The energy of the LOB is indeed similar to the typical energy of a point defect in bcc ^{4}He measured by different techniques.[18,19] A neutron incident on the solid can create a vacancy by knocking a single atom away from its lattice site. However, single particle excitations have a large energy width, because the final state of the atom is in the continuum.[4] Hence, this possibility is not consistent with the small linewidth of the LOB. The same argument applies to the creation of vacancy-interstitial pairs (Frenkel pairs). The formation energy of a Frenkel pair depends on the interstitial-vacancy distance, which can take different values. Thus, one expects a broad feature in the scattering intensity vs energy rather than the narrow peak, which is observed (Fig. 3). In addition, recent simulations of point defects in bcc He indicate that a vacancy branch has a considerable dispersion.[20] This result supports our claim that the dispersionless LOB branch is probably not associated with the creation of vacancies. Another possibility is that the neutrons excite some resonant mode,[17,21] namely, internal vibrations of split interstitials or vacancies already present in the crystal. Formation energies and resonant mode energies of point defects should be similar, and in addition, the energy width of the resonant modes is small. Some qualitative evidence of the presence of such excitations was observed in our simulations of crystals containing a large number of interstitials.[13] Hence, although we did not find any analytical calculations of these modes for solid He, this possibility is of interest. Finally, a feature similar to the LOB may have been observed in neutron scattering from the hcp solid phase at a temperature of 100 mK.[22] Resonant modes, if they exist, should be observed in both hcp and bcc He. Further investigation of these features in both solid phases may help to understand their origin.

In conclusion, we investigated the recently reported opticlike excitation branch HOB, and found it to be anisotropic and weakly dispersive. In addition, another excitation branch was discovered (LOB). This branch is also anisotropic, dispersionless, and with a very small linewidth. In contrast with the HOB, the LOB does not couple to phonons. One possible interpretation is that these branches are associated with point defects or excitations thereof. However, none of the existing models is detailed enough to allow meaningful comparison with the data.

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*Electronic address: poshri@tx.technion.ac.il; URL: http://physics.technion.ac.il/~poshri

†Electronic address: emilp@physics.technion.ac.il; URL: http://physics.technion.ac.il/~emilp/

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