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Measurement of acoustic phonons in UBe₁₃

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The low-energy phonon dispersion of UBe₁₃ has been measured by means of inelastic neutron scattering at room temperature and 10 K and elastic constants have been extracted. They are consistent with values obtained by scaling from pure beryllium, and the angular variation is almost isotropic. The lattice contribution to the low-temperature specific heat has been calculated from these data and is compared with the measured specific heats in the literature.

I. INTRODUCTION

Although the intermetallic compound UBe₁₃ was first produced and its structure determined over thirty years ago,^{1,2} interest in it has recently increased dramatically following the discovery that it exhibits bulk superconductivity and that the electrons responsible have very high effective masses.³ A crucial piece of evidence in this discovery has been the temperature variation of the specific heat.^{3,4} While the major interest has been in the electronic component, it is necessary to subtract off the lattice component C_{latt} , which is normally assumed to obey the T^3 Debye law at low temperatures. The best way to determine the coefficient of this term is to measure the sound velocities by ultrasonic or neutron scattering methods and then perform a surface integral over the low-energy states:

$$C_{\text{latt}} = AT^3, \quad (1)$$

where

$$A = \frac{12\pi^4 n N_a k}{5\theta_D^3} = \frac{2\pi^2 k^4 N_a v_0 \langle v^{-3} \rangle}{5\hbar^3},$$

where N_a is Avogadro's number, v_0 is the volume of one formula unit, θ_D is the Debye temperature, and the angular average of the sound velocity is given by

$$\langle v^{-3} \rangle = \frac{1}{3} \sum_{i=1}^3 \int v_i^{-3} \frac{d\Omega}{4\pi}. \quad (2)$$

Although some ultrasonic measurements have been made⁵ at low temperatures, accurate values for elastic constants have yet to be reported. In this paper, we report elastic constants as determined by means of inelastic neutron scattering at ambient temperature and at 10 K.

The lattice is face-centered cubic,¹ with a basis of 28 atoms (2 formula units) and there are 84 independent normal modes. We have only attempted to measure the low-energy part of the spectrum, in order to obtain reliable elastic constants from the three acoustic modes. However, it is to be expected that some optic modes, due to vibrations of the uranium atoms against the more rigid beryllium lattice, will be quite low in energy. In fact, these have recently been observed in a polycrystalline sample on a time-of-flight neutron spectrometer⁶ and have been found to lie at approximately 13 meV. The fact that they lie at this energy means that they make no contribution to the low-temperature lattice specific heat, which is dominated by the acoustic modes.

II. RESULTS

The sample used in this study was a 0.38 cm³ single crystal grown in an Al flux as reported previously.³ The experiments were performed on the H7 triple-axis spectrometer at the Brookhaven National Laboratory High-Flux Beam Reactor. Pyrolytic graphite was used as monochromator and analyzer and graphite filters were used to reduce higher-

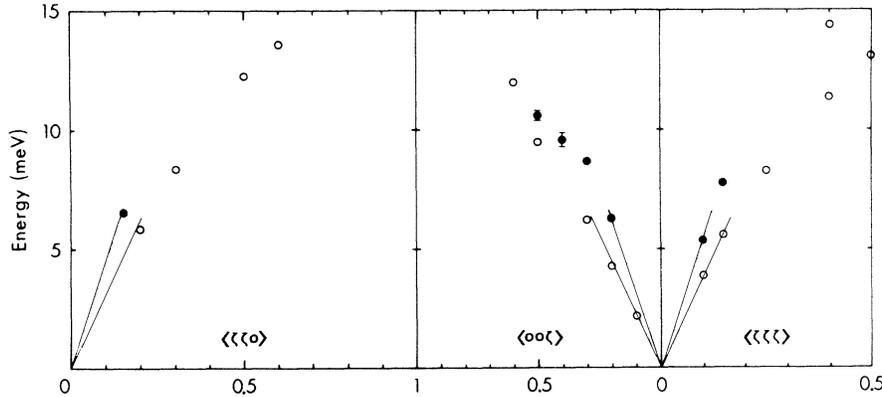


FIG. 1. Phonon energies along symmetry directions in UBe_{13} at room temperature. The solid circles represent longitudinal phonons and the open circles transverse phonons. The straight lines have gradients corresponding to sound velocities calculated from the room-temperature elastic constants listed in Table I.

order contamination. The values of collimation used were $20^\circ\text{-}20^\circ\text{-}20^\circ\text{-}20^\circ$ and $20^\circ\text{-}20^\circ\text{-}20^\circ\text{-}40^\circ$. The crystal was aligned with the $\langle 1\bar{1}0 \rangle$ axis perpendicular to the scattering plane and measurements were made along the $\langle 001 \rangle$, $\langle 110 \rangle$, and $\langle 111 \rangle$ symmetry directions. We were able to measure all of the acoustic modes in those directions, except for the $\langle 110 \rangle$ transverse mode polarized perpendicular to the scattering plane, with velocity $\sqrt{(c_{11} - c_{12})/2\rho}$. Our room-temperature results are shown in Fig. 1. These data have been corrected for instrumental resolution, and dispersion was observed in all branches for which more than one measurement was made. Sound velocities were extracted by extrapolating back to $\zeta = 0$, assuming that the phonon frequencies vary as

$$\omega = \nu q - \beta q^3 \quad (3)$$

for small wave vector q in any given direction and that higher-order terms are negligible. These, in turn, have been fitted to the three independent elastic constants c_{11} , c_{12} , and c_{44} . The values so obtained are listed in Table I and the sloping lines in Fig. 1 have gradients corresponding to velocities calculated using these elastic constants. Similar, though slightly less extensive, measurements were made at 10 K and the elastic constants, obtained in the same manner, are also listed in Table I. There is clearly some hardening of the lattice as the temperature is lowered (by $\sim 10\%$, which is much greater than would be expected from the change in lattice parameter). At both temperatures, the elastic constant c_{12} is very small and may even be negative. This is consistent with recent measurements⁷ of Poisson's ratio in UBe_{13} , which indicate that c_{12} is small and positive.

TABLE I. Elastic constants of UBe_{13} in 10^9 Nm^{-2} at room temperature and 10 K.

| | Room temperature | 10 K |
|----------|------------------|-------------|
| C_{11} | 265 ± 16 | 310 ± 9 |
| C_{12} | 7 ± 22 | -1 ± 9 |
| C_{44} | 134 ± 3 | 161 ± 5 |

While it is not clear what physical significance this has, we note that large negative values of c_{12} have been observed in a number of intermediate-valent compounds.^{8,9} In no cases were the measured phonon groups broader than the instrumental resolution. We therefore have no evidence for strong coupling between phonons and the "heavy fermions" within the energy range of our experiment.

In the course of these measurements, while studying longitudinal modes in the vicinity of the $(4,4,4)$ and $(6,0,0)$ reciprocal lattice points, we have observed peaks at energies lower than those of the longitudinal modes. They occur at energies very much like those of transverse modes of the same wave vector, while the $(\mathbf{Q} \cdot \mathbf{e})$ polarization factor in the cross section is nominally zero for transverse modes measured in this manner. However, our resolution calculations indicate that this intensity can be accounted for as entirely due to transverse modes picked up slightly off axis, but still within the finite resolution of the spectrometer.

III. DISCUSSION

The expression in Eq. (2) has been evaluated for cubic crystals that are nearly isotropic and the resultant expression is^{10,11}

$$\langle v^{-3} \rangle = \frac{\rho^{3/2}}{3} \left[\frac{2}{c_{44}^{3/2}} + \frac{1}{c_{11}^{3/2}} + \frac{3}{5} (c_{12} - c_{11} + 2c_{44}) \left(\frac{1}{c_{44}^{5/2}} - \frac{1}{c_{11}^{5/2}} \right) \right] \quad (4)$$

Clearly, we only need to substitute the values for the elastic constants, listed in Table I, into Eqs. (1), (2), and (4) to obtain the coefficient of the cubic term in the low-temperature lattice specific heat. When this is done we obtain a value of $A = 10.49 (\pm 0.36) \times 10^{-5} \text{ J mole}^{-1} \text{ K}^{-4}$ which corresponds to $\theta_D = 638 \pm 8 \text{ K}$. The errors quoted here were obtained solely by propagating the original statistical uncertainties in the observed neutron groups and the agreement with the Debye temperature ($\sim 620 \text{ K}$) extracted from the specific-heat measurements⁴ is very good.

UBe_{13} is 93% beryllium and the beryllium atoms are grouped together into 12-fold coordinated clusters, a local

environment which is similar to the hexagonal close-packed structure of pure beryllium. Furthermore, the Be-Be nearest-neighbor distances range between 2.154 and 2.256 Å, while that in pure beryllium is 2.286 Å. It is therefore reasonable to suppose that the elastic behavior of the two materials will be closely related. Beryllium itself is fairly isotropic,^{12,13} with a longitudinal sound velocity v_L ranging between 12700 and 13600 ms⁻¹ and a transverse sound velocity v_T ranging between 8500 and 9500 ms⁻¹ at room temperature. The greatest difference between the two materials is the introduction of the massive uranium atom into the lattice. On changing from UBe₁₃ to an imagined isostructural BeBe₁₃, the sound velocities would change in proportion to the inverse square root of the mass of a formula unit, provided that the interatomic force constants and spacings remain unchanged. The mass of a formula unit is greater by a factor of 2.82. However, pure beryllium and BeBe₁₃ are not the same. The next effect to consider is that of changing the average atomic volume. Ignoring the effects of the detailed atomic arrangement on the force constants, it is plausible (from a consideration of a simple ball and spring model) that the sound velocity will vary as the cube root of the atomic volume. Within these assumptions, which are essentially the same as those used by Overhauser and Appel,¹⁴ we can then make a quantitative comparison between UBe₁₃ and pure beryllium. The average atomic volume of UBe₁₃ is 19% greater than in pure beryllium, so the UBe₁₃ sound velocities should be 0.631 of the beryllium sound velocities. Scaling the room-temperature values of Smith and Abrogost,¹² we predict that $v_T = 5360\text{--}6000$ ms⁻¹ and $v_L = 8010\text{--}8580$ ms⁻¹, while the values of the room-temperature elastic constants in Table I give $v_T = 5440\text{--}5510$ ms⁻¹ and $v_L = 7760\text{--}7850$ ms⁻¹. The agreement is good.

Of course, the interatomic force constants are unlikely to

remain unchanged. For a simple uniform expansion of the pure beryllium lattice, one can estimate the magnitude of this effect using the Grüneisen constant γ , which is a simple measure of anharmonicity and relates changes in phonon frequency to volume changes:

$$\frac{\Delta\omega}{\omega} = -\gamma \frac{\Delta V}{V} \quad (5)$$

Using standard thermodynamic relations¹⁵ and published values of the bulk modulus, linear coefficient of thermal expansion and specific heat, we obtain $\gamma = 1.14$ for beryllium. This is very low compared with other materials. Even so, this would then lead to a further 22% softening of the sound velocities, in addition to the effects described in the previous paragraph. This is at variance with our data, indicating that the electron gas cannot be thought of as simply dilated compared with beryllium. In fact, it may be more appropriate to consider the volume of the 12-fold coordinated beryllium cluster, which is compressed compared with pure beryllium, as the appropriate parameter. In any case, the basic notion of a direct comparison with pure beryllium works well as a first approximation.

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