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Soft phonons in superconducting $LuNi_2B_2C$

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Inelastic-neutron-scattering techniques have been used to measure the low-lying phonon-dispersion curves of superconducting $LuNi_2B_2C$ along the $[\xi 00]$ and $[00\xi]$ symmetry directions. The most important result of this experiment is that the phonon frequencies of the acoustic and first optical Δ_4 $[\xi 00]$ branches in the vicinity of the zone-boundary point G_1 decrease with decreasing temperature. Actually these two branches exhibit pronounced dips at low temperatures, close to G_1. This shows that the electron-phonon interaction is quite strong and causes an incipient lattice instability, a behavior typical of strongly coupled conventional superconductors. Furthermore, the phonon anomalies occur at wave vectors close to those of the incommensurate magnetically ordered structures observed in the magnetic compounds of this family, which suggests that both the magnetic ordering and the incipient lattice instabilities are influenced by common nesting features of the Fermi surfaces of the rare-earth nickel boride carbides.

I. INTRODUCTION

The quaternary intermetallic compounds of the recently discovered[1–4] family of rare-earth nickel boride carbides, RNi_2B_2C (where R stands for a rare-earth element), have very interesting physical properties. The structure[3] of these compounds is body-centered tetragonal (space group $I4/mmm$) and consists of R-C layers separated by Ni_2B_2 sheets. Many of these compounds are superconducting with the highest superconducting temperatures, 16.6 and 15.6 K, observed[2–5] for the Lu and Y compounds, respectively. Particularly interesting is that superconductivity is not only observed for nonmagnetic rare-earth elements but also for magnetic rare-earth elements such as Tm, Er, Ho,[2–5] and most recently Dy;[6,7] the Tm, Ho, Er, and Dy compounds are superconducting with superconducting temperatures of 10.8, 8, 10.5, and 6.2 K, respectively, whereas the Gd and Tb compounds are not superconducting at least down to approximately 2 K. This family of intermetallic compounds is, therefore, particularly well suited for a detailed study of the interplay between superconductivity and magnetism, which has been previously examined[8–11] in the RRh_4B_4 and RMo_6S_8 magnetic superconductors.

The magnetic structure of Ho, Er, and Dy compounds have been studied[7,12–15] by neutron-diffraction techniques and that of $GdNi_2B_2C$ by resonant magnetic x-ray scattering.[16] In $DyNi_2B_2C$ and below 4.7 K in $HoNi_2B_2C$ a simple antiferromagnetic structure, consisting of ferromagnetically aligned basal-plane layers with the magnetic moments of two consecutive layers pointing in opposite directions, has been observed. In the Ho compound, two incommensurate modulations of the moments were observed between 4.7 and 6 K, one with wave vector along $\mathbf{c}^*$, and the other with wave vector along $\mathbf{a}^*$. The latter modulation has also been observed in the Er and Gd compounds. The above experimental results suggest[7,12–15] that there are common Fermi surface nesting features along $\mathbf{a}^*$ and/or $\mathbf{c}^*$ which cause the magnetic ordering of the rare-earth magnetic moments via the Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism.

Electronic band-structure calculations[17–19] suggest that these materials are conventional superconductors with a relatively high density of states at the Fermi level. These calculations show that there is a rather complex set of bands crossing E_F which are strongly coupled to the phonons and may be responsible for the superconducting properties of these compounds. More recently, Mattheiss, Siegrist, and Cava suggested[20] that superconductivity in these systems can be attributed to a conventional electron-phonon mechanism that couples the sp-like conduction electrons to a high-frequency boron A_{1g} phonon mode in which the boron atoms move along the c axis relative to the other atoms. The energy of the Raman active boron A_{1g} mode was calculated[17] by Pickett and Singh to be 106 meV($=850$ cm^{-1}), a value in good agreement with Raman-scattering measurements.[21,22] Furthermore, Pickett and Singh obtained an estimate of 20 meV for the average phonon frequency by using resistivity data to estimate the electron-phonon coupling parameter ($\lambda \simeq 2.6$). Since this estimate of an average phonon energy is considerably lower than the energy of the boron A_{1g} mode, they suggest that superconductivity in these compounds may involve soft modes or strong contributions from the heavier atoms.

More recently, Rhee, Wang, and Harmon[23] performed a calculation (without matrix elements) of the generalized electronic susceptibility $\chi(q)$ of $LuNi_2B_2C$ based on the normal-state electronic band structure of this compound. Peaks in the calculated $\chi(q)$ occur near wave vectors corresponding to those observed for the incommensurate magnetic structures observed in $HoNi_2B_2C$. Of particular interest is that the sharp peak in the calculated $\chi(q)$ along $\mathbf{a}^*$ is due to strong Fermi surface nesting and occurs at a wave vector of approximately 0.6, which is close to the values of the incommensurate magnetic modulations along $\mathbf{a}^*$ observed in the Ho, Er, and Gd compounds (0.585, 0.553, and 0.553, respectively). If such a strong nesting feature is indeed present at the Fermi surface of $LuNi_2B_2C$, one also would expect strong Kohn anomalies in the phonon dispersion curves of this compound.

It is clear from the above discussion that experimental

studies of the lattice dynamical properties of these compounds may lead to a better understanding of their properties. No detailed experimental study of the phonon dispersion curves of these compounds has been performed to date, since the presently available crystals are not of sufficient size for inelastic-neutron-scattering experiments. However, by mounting two crystals together, we were able to measure several low-lying phonon dispersion curves of the Lu compound. In this report we present the results of these experiments.

II. EXPERIMENT

Single crystals of $LuNi_2B_2C$ were grown at the Ames Laboratory by the high-temperature flux technique as described elsewhere.[24] The crystals obtained are platelets with the c axis perpendicular to their flat surface. Since the size of the crystals (approximately $5\times5\times0.5$ mm^3) is relatively small for inelastic neutron scattering, a composite crystal, consisting of two of these crystals, was used in the present experiments. The two crystals were mounted together and their alignment was adjusted until their Bragg reflections were found to coincide to within the instrumental resolution.

The experiments were performed using the HB1A, HB2, and HB3 triple-axis spectrometers at the high flux isotope reactor of the Oak Ridge National Laboratory. Pyrolitic graphite reflecting from the (002) planes was used as both monochromator and analyzer in the HB2 and HB3 spectrometers and the data were collected using fixed-scattered-neutron energies of 14.7 and 30.5 meV. The HB1A spectrometer is a constant incident energy (14.7 meV) instrument utilizing a double pyrolitic graphite monochromator [reflecting from the (002) planes]. The collimation before the sample was either 20 or 40 min of arc and that after the sample was 40 min of arc. Pyrolitic graphite filters were used in all spectrometers to attenuate higher-order contaminations.

All data were collected with the crystal oriented so that the scattering plane coincides with the a-c crystal plane. Measurements of the phonon dispersion curves were performed along the $[\xi00]$ and $[00\xi]$ high symmetry directions. The measurements along the $[\xi00]$ direction were extended beyond the zone-boundary point G_1 [at $(\pi/a)(1+a^2/c^2)$] to $(2\pi/a,0,0)$ which is the same point as the zone-boundary point Z along the $[00\xi]$ direction. While most measurements were performed at room temperature, the temperature dependence of the phonon frequencies of the interesting Δ_4 branches along the $[\xi00]$ direction was determined by measurements at 120, 60, 25, 10, and 2 K. A selected number of phonon frequencies of the other branches were also measured at 10 K to assess whether they exhibit anomalous behavior.

The measured frequencies were assigned to the various branches by comparing the measured intensities of the corresponding neutron groups with calculations of intensities based on Born–von Kármán force-constant models; the parameters of the force-constant models were determined by fitting the neutron-scattering data along with the frequencies of the measured[21,22] Raman-active modes. The way we have chosen to satisfy the compatibility relations at the zone-boundary point Z is only one of several alternatives, since the phonon frequencies belonging to different representations

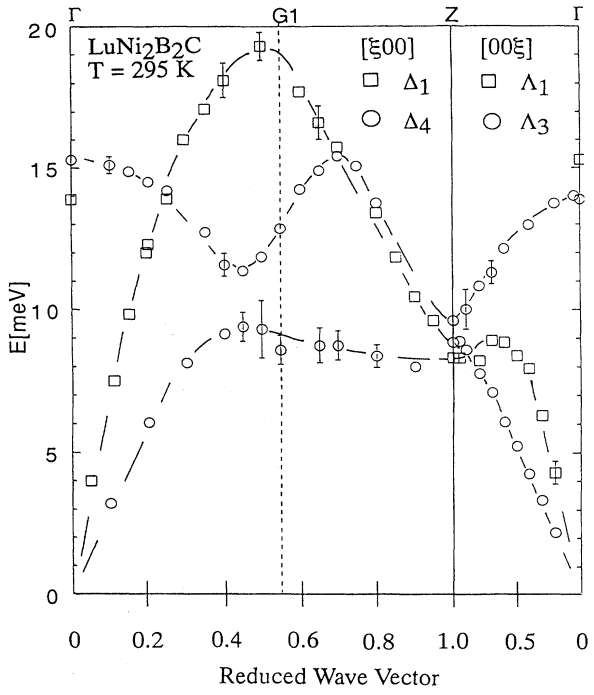

FIG. 1. Room-temperature acoustic and lowest-lying optical phonon dispersion curves of $LuNi_2B_2C$ along the $[\xi00]$ and $[00\xi]$ symmetry directions. The lines are intended as guides to the eye. The size of the symbols is a measure of the estimated uncertainties in the measured frequencies.

are so close to each other in the vicinity of Z that the corresponding neutron groups cannot be sufficiently resolved with the resolution of the present experiment.

The lowest lying acoustic and optical branches were both obtained by measurements from the (008) reciprocal point, i.e., with a configuration appropriate for transverse branches with polarization along the c axis. By symmetry, the acoustic branch with this polarization belongs to the Δ_4 representation. Based on the analysis of the data using a Born–von Kármán force-constant model (see previous paragraph), the optical branch was also assigned to the Δ_4 $[\xi00]$ representation. For small wave vectors, both Δ_4 branches are purely transverse with atomic displacements along the c axis. For the acoustic branch all atoms move in phase, whereas in the optical branch the motion of the Ho atom is out of phase with that of the other atoms. For larger values of the wave vector the Ho and C displacements remain transverse, while the Ni and B atoms develop a longitudinal component.

III. RESULTS AND DISCUSSION

At room temperature, the phonon frequencies of the acoustic and optical branches, with zone-center energies at approximately 14, 15, and 24 meV, were measured. The room-temperature acoustic and lowest lying optical branches are plotted in Fig. 1. The highest measured energy at the zone center ($\simeq$24 meV) is in good agreement with the experimentally determined[21,22] lowest Raman-active Ni-B_{1g} mode. A complete analysis of these measurements in terms of lattice dynamical models will be published elsewhere.

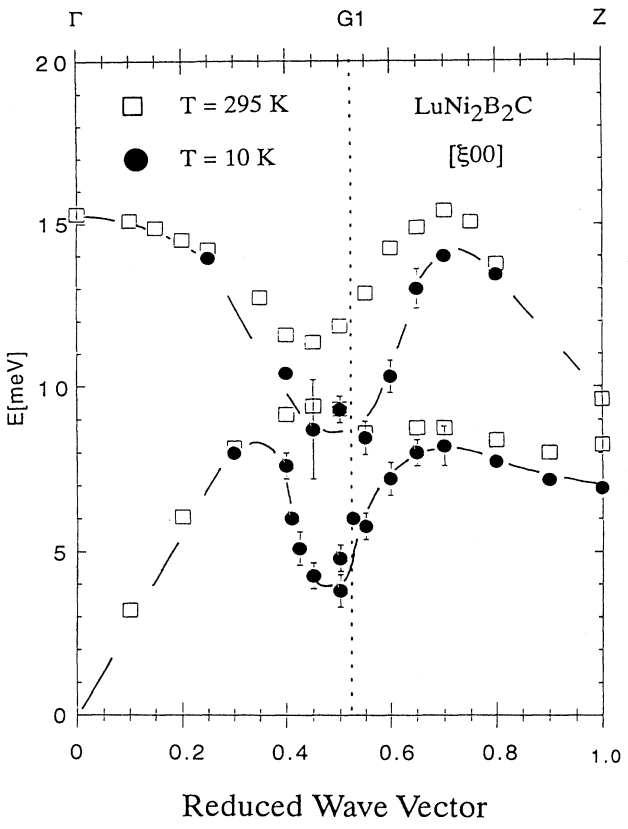

FIG. 2. The Δ_4 [ξ00] branches at 295 and 10 K. The lines through the 10 K points are intended as guides to the eye.

The most interesting result of these experiments is the temperature dependence of the observed phonon frequencies. The frequencies of selected phonons on all the measured branches, except the Δ_4 [ξ00], show no significant change with temperature. The phonon frequencies of the acoustic and first optical Δ_4 [ξ00] branches in the vicinity of the zone-boundary point G_1, on the other hand, decrease with decreasing temperature. Actually, these two branches exhibit pronounced dips at low temperatures close to G_1 (Fig. 2). This is quite a unique case of soft phonon behavior: the softening occurs over the same region of wave vectors for two (Δ_4) branches that cannot cross by symmetry.

The observed phonon softening shows that the electron-phonon interaction is quite strong in this compound and causes an incipient lattice instability, a behavior which is typical of conventional superconductors with relatively high-superconducting transition temperatures. This observation provides experimental support for the argument, based on band theoretical calculations,[17–19] that this compound is a conventional superconductor. The strong electron-phonon interaction is presumably responsible both for the relatively high T_c of this compound as well as the observed incipient lattice instability.

As mentioned in the Introduction, Pickett and Singh argued[17] that soft phonon modes, as those observed in this experiment, may be involved in determining the superconducting properties of this compound. However, the importance of the observed soft phonons in determining T_c relative to that of the boron A_{1g} mode in the model[20] of Mattheis, Siegrist, and Cava cannot be presently ascertained without comparing the experimental results with detailed lattice dynamical calculations incorporating the electron phonon interaction.

The calculated[23] (without matrix elements) generalized electronic susceptibility of $LuNi_2B_2C$ along [ξ00] exhibits a pronounced peak close to G_1 due to a strong nesting feature of the Fermi surface of this compound. This nesting feature may be responsible for the observed incommensurate magnetic ordering wave vectors in the magnetic rare-earth nickel boride carbides. The same Fermi surface nesting feature may be responsible for the strong anomalies observed in the present experiment for the Δ_4 [ξ00] phonon modes close to G_1. Therefore, it appears that in the magnetic compounds of this family we may have the interesting situation where, due to the nesting of the Fermi surface, phonon softening and magnetic ordering are competing to decrease the energy of the system. Therefore, the electron-phonon and RKKY interactions in the magnetic superconducting systems are extremely interesting and further experimental and theoretical investigations will be required to establish a detailed understanding of the low-temperature properties of these materials. An experimental study of the phonon dispersion curves of $HoNi_2B_2C$ is presently in progress in this laboratory.

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