

Temperature dependence of low-energy phonons in magnetic nonsuperconducting TbNi₂B₂CS. Anissimova,¹ A. Kreyssig,^{2,6} O. Stockert,⁴ M. Loewenhaupt,² and D. Reznik^{1,3,5}¹*Department of Physics, University of Colorado at Boulder, Boulder, Colorado 80309, USA*²*Technische Universität Dresden, Institut für Festkörperphysik, D-01062 Dresden, Germany*³*CEA Saclay, Laboratoire Léon Brillouin, F-91191 Gif sur Yvette, France*⁴*Max-Planck-Institut für Chemische Physik fester Stoffe, Nöthnitzer Straße 40, D-01187 Dresden, Germany*⁵*Karlsruher Institut für Technologie (KIT), Institute für Festkörperphysik, Postfach 3640, D-76121 Karlsruhe, Germany*⁶*Ames Laboratory and Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011, USA*

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We report temperature dependence of low-energy phonons in magnetic nonsuperconducting TbNi₂B₂C single crystals measured by inelastic neutron scattering. We observed low-temperature softening and broadening of two phonon branches, qualitatively similar to that previously reported for superconducting RNi₂B₂C ($R =$ rare earth, Y) compounds. This result suggests that superconductivity in TbNi₂B₂C compounds is absent not because of weak electron-phonon coupling but as a result of pair breaking due to magnetism.

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I. INTRODUCTION

In some rare-earth nickel borocarbides RNi₂B₂C, superconductivity coexists with magnetic order.^{1,2} Extensive neutron and x-ray scattering experiments revealed an incommensurate magnetic structure below approximately 15 K in both superconducting Er, Ho³⁻⁶ and nonsuperconducting Tb, Gd^{7,8} compounds. This observation was interpreted in terms of common Fermi-surface nesting features along a^* , which cause magnetic ordering of the rare-earth moments via the Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism.⁷ ⁵⁷Fe Mössbauer spectroscopy and muon-spin relaxation (μ SR) studies of polycrystalline TbNi₂B₂C (Ref. 9) confirmed the presence of a small ferromagnetic component below about 8 K previously observed via neutron diffraction⁷ and magnetization measurements.^{10,11}

In addition to magnetic effects, strong phonon softening has been observed in superconducting RNi₂B₂C single crystals with $R =$ Lu, Y, Er, and Ho,¹²⁻¹⁵ while no significant temperature dependence of the phonon spectra was detected for the nonsuperconducting TbNi₂B₂C (Ref. 16). The superconducting transition temperature T_c systematically decreases for RNi₂B₂C ($R =$ Lu, Y, Tm, Er, and Ho) upon going from Lu ($T_c = 16.6$ K) to Ho ($T_c = 7.5$ K). This observation was interpreted by H. Eisaki *et al.* in terms of increasing coupling between the rare-earth magnetic moments and the conduction electrons,¹ which suppressed superconductivity. For TbNi₂B₂C, this pair breaking could be strong enough to completely destroy superconductivity. An alternative possibility for the absence of superconductivity in this system is that electron-phonon coupling is weaker than in the superconducting compounds.¹⁶

We investigated the strength of electron-phonon coupling in TbNi₂B₂C by detailed measurements of the temperature dependence of low-energy phonons in the magnetic nonsuperconducting TbNi₂B₂C by inelastic neutron scattering. The observed softening of these phonon branches in this compound upon cooling from 300 to 30 K indicates that electron-phonon interactions in magnetic nonsuperconducting TbNi₂B₂C are strong and, although weaker, are of the same order of magnitude as those in superconducting RNi₂B₂C compounds.

II. EXPERIMENTAL DETAILS

Intermetallic RNi₂B₂C borocarbides crystallize in the body-centered tetragonal LuNi₂B₂C structure with the space group $I4/mmm$, which consists of R -C layers separated by Ni₂B₂ sheets.¹⁷ All crystallographic parameters, i.e., the lattice constants a and c as well as the z position of boron, scale roughly with the ionic radius of the rare earth. TbNi₂B₂C compounds exhibit antiferromagnetic ordering at low temperatures below 15 K with the magnetic Tb³⁺ moments aligned along the a axis and modulated with the propagation vector $\tau = (0.545, 0, 0)$ (Refs. 7 and 18). No superconductivity has been detected in this compound down to 7 mK.¹⁹

Rodlike TbNi₂B₂C single crystals were grown by the floating zone method using ¹¹B isotope to avoid strong neutron absorption.²⁰ Specimens with a length of 6 mm were cut from the rods with 6 mm diameter. For the inelastic neutron-scattering experiments the samples were oriented in the a - c scattering plane. The measurements were performed on the triple-axis spectrometer 1T1 at the Laboratoire Léon Brillouin, Saclay. Phonons propagating in the $[\xi 0 0]$ direction, where ξ is a reduced wave vector, were recorded in the Brillouin zone centered at $(0, 0, 8)$ by energy scans using a fixed final energy $E_f = 14.8$ meV at temperatures between $T = 2$ and 300 K.

III. EXPERIMENTAL RESULTS

When phonons weakly couple to the conduction electrons, their energies and lifetimes slightly harden and narrow upon cooling due to decreased anharmonicity. However, when phonons strongly couple to conduction electrons in the nearly nested regions of the Fermi surface, they soften and broaden upon cooling as the Fermi surface sharpens. Inelastic neutron- or x-ray-scattering measurements of phonons are the most direct way to identify the phonon modes strongly coupled to electrons. Due to the relatively large size of the available single crystal, we carried out our investigation using inelastic neutron scattering.

Figure 1 illustrates how we determined the background that was subtracted from the raw data to obtain the phonon spectra. It shows energy scans near the zone center at $T = 30$ K.

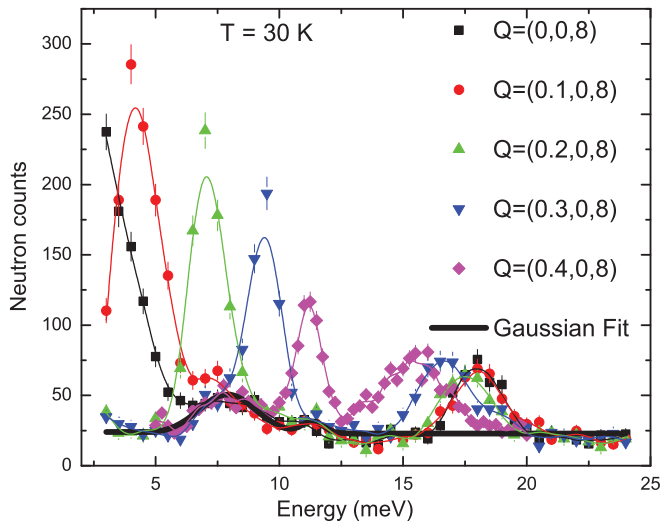


FIG. 1. (Color online) The $\xi = 0$, $\xi = 0.1$, $\xi = 0.2$, $\xi = 0.3$, and $\xi = 0.4$ spectra at $T = 30$ K. The bold solid line represents the crystal electric-field excitations fit with a Gaussian line shape. Solid lines represent smoothed data.

The strongest peaks result from upward-dispersing acoustic phonons. The inelastic peaks between 15 and 18 meV originate from downward-dispersing optical phonons. In addition to phonons, crystal-electric-field (CEF) transitions are present between 5 and 13 meV.²¹ For the purpose of studying the phonons, the CEF excitations contribute to the background. CEF excitations are dispersionless, and their form factor is nearly Q independent in the narrow Q range of interest. We determined them from the spectra where they do not overlap with phonons. The bold solid line in Fig. 1 represents the background, which includes Gaussian peaks due to CEF excitations and a straight line due to other energy-independent sources of background. It was subtracted from the data to isolate the one-phonon scattering shown in subsequent figures.

Figure 2 shows the temperature dependencies of phonon spectra at different wave-vector transfers (Q) to the neutron at 30 and 300 K. The CEF contribution was subtracted from all low-temperature spectra as described above. We did not attempt to identify the contribution of the CEF excitations at 300 K, because they become weaker at high T , whereas phonons become stronger due to the Bose factor, $n = 1/(1 - e^{-\hbar\omega/k_B T})$. Thus their contribution to the scattering intensity becomes negligible. The data were divided by the Bose factor to correct for the temperature dependence of the one-phonon scattering, which also removed most temperature dependence from the background. High-temperature curves were scaled with the low-temperature ones by subtracting a constant, which is reasonable, because the background increases with T . Phonon spectra of the optical mode between 10 and 15 meV soften and broaden from 300 to 30 K at $Q = (0.5,0,8)$, $Q = (0.6,0,8)$, $Q = (0.7,0,8)$, and $Q = (0.8,0,8)$, whereas the spectra at $Q = (0.4,0,8)$, $Q = (0.9,0,8)$ (not shown), and $Q = (1,0,8)$ are nearly temperature independent.

Figure 3 shows the dispersion of the two low-energy acoustic and optical phonon branches propagating along the $[\xi 0 0]$ direction at $T = 30$ and 300 K. The energy scans were

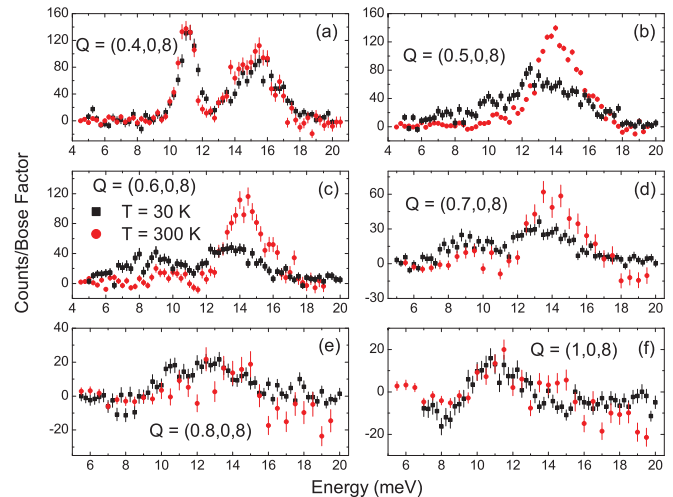


FIG. 2. (Color online) Background subtracted phonon spectra divided by the Bose factor taken at $\xi = 0.4$ (a), $\xi = 0.5$ (b), $\xi = 0.6$ (c), $\xi = 0.7$ (d), $\xi = 0.8$ (e), and $\xi = 1$ (f) at $T = 30$ K (black squares) and 300 K (red circles).

fitted by Gaussian peaks. The dispersion of the two interesting phonon branches at $T = 2$ K is similar to the dispersion measured in the temperature range $T = 11$ –100 K, and there is no softening below 100 K.¹⁶ However, strong softening of both branches appears between 100 and 300 K between $Q = (0.5,0,8)$ and $(0.75,0,8)$. Our low-temperature data reveal a discrepancy in the energy range $\hbar\omega = 8$ –10.5 meV as compared to Ref. 16, which can be attributed to a different interpretation of CEF contribution. Unlike Ref. 16, we observe a strong dip of the low-energy phonon branch for wave vectors between $Q = (0.5,0,8)$ and $(0.75,0,8)$ at low temperatures, which is weaker but of the same order of magnitude as the one found in superconducting $\text{HoNi}_2\text{B}_2\text{C}$. This result indicates that electron-phonon coupling in $\text{TbNi}_2\text{B}_2\text{C}$ is strong enough

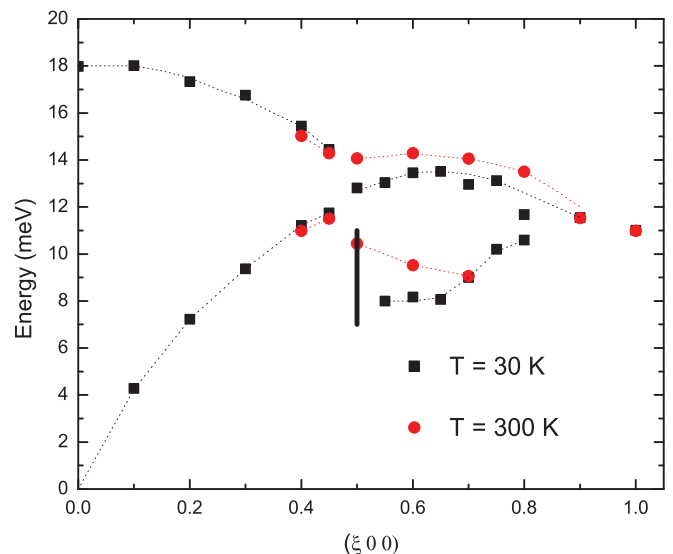


FIG. 3. (Color online) Low-energy phonons along $[\xi 0 0]$ at 30 and 300 K. The dashed lines are guides for the eye. The vertical black bar at $\xi = 0.5$ denotes a broad phonon peak at 30 K containing also contributions from the next upper phonon branch.

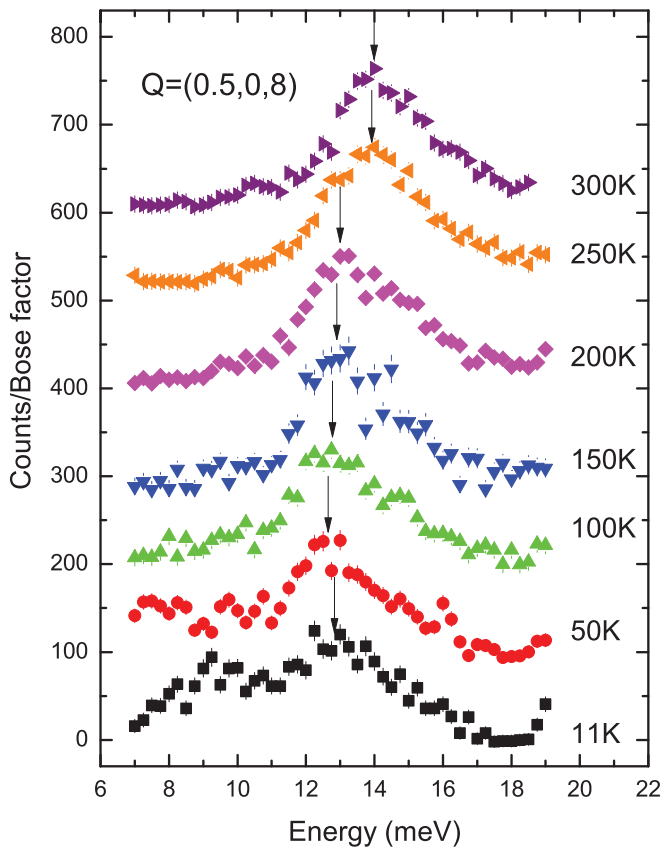


FIG. 4. (Color online) Phonon spectra divided by the Bose occupation factor for a wave vector $(0.5, 0, 8)$ in the temperature range from 11 to 300 K. The curves are vertically shifted for clarity. The phonon-peak position is indicated by arrows.

to mediate superconductivity (perhaps with a lower T_c) in the absence of a pair-breaking mechanism.

Figure 4 illustrates the temperature dependence of the optic phonon at $Q = (0.5, 0, 8)$. Here we show the raw data divided by the Bose factor without subtracting the background. The curves are vertically shifted with the arrows indicating the phonon peak position. The lowest temperature scan contains an additional feature at 9 meV to which both the acoustic phonon and the CEF excitation contribute. We have not measured the temperature dependence of the CEF excitation, so it is

not possible to determine the temperature dependence of the acoustic phonon based on the available data. However, the optic phonon does not overlap with the CEF excitation, and the data on its T dependence are unambiguous. We observe a softening from room temperature to $T \cong 100$ K of 1 meV. Below ≈ 100 K, it shows no peak position shift.

IV. DISCUSSION AND CONCLUSIONS

Strong softening of two low-energy phonon branches was observed by neutron scattering in superconducting RNi_2B_2C single crystals with $R = Lu, Y,$ and Er .^{12–15} Point-contact spectroscopy revealed strong electron-phonon interaction in the superconducting RNi_2B_2C compounds with $R = Y$ and Ho , in contrast to the nonsuperconducting $LaNi_2B_2C$,²² which suggests that the presence of superconductivity is controlled by the strength of electron-phonon coupling as opposed to magnetic pair breaking. However, our inelastic neutron scattering experiments on nonsuperconducting $TbNi_2B_2C$ clearly demonstrate strong electron-phonon coupling in this compound. Thus we conclude that superconductivity in $TbNi_2B_2C$ is absent due to magnetic pair breaking.

Previous studies of the competition between magnetism and superconductivity in RNi_2B_2C superconductors with the magnetic rare-earth elements $R = Lu, Tm,$ and Er revealed a very weak coupling between the rare-earth magnetic moments and the conduction electrons due to a small conduction-electron density at the rare-earth site.¹ In contrast, a strong magnetic pair-breaking effect has been observed in Dy and Tb samples¹ which gives additional evidence that superconductivity in magnetic $TbNi_2B_2C$ is indeed destroyed by the substantially strong interaction between the local magnetic moments and the conduction electrons. In summary, electron-phonon coupling in magnetic nonsuperconducting $TbNi_2B_2C$ is strong enough to mediate superconductivity. This implies that its absence can only result from magnetic pair breaking.

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