Temperature dependence of low-energy phonons in magnetic non-superconducting TbNi$_2$B$_2$C

S. Anissimova$^1$, A. Kreyssig$^{2,6}$, O. Stockert$^4$, M. Loewenhaupt$^2$, D. Reznik$^{1,3,5}$

$^1$Department of Physics, University of Colorado at Boulder, Boulder, Colorado 80309
$^2$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 TbNi$_2$B$_2$C 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 nonsuperconducting RNi$_2$B$_2$C ($R$ = rare earth, $Y$) compounds. This result suggests that superconductivity in TbNi$_2$B$_2$C 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 $RNi_2B_2C$, 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, Ho [3–5] and nonsuperconducting Tb, Gd [6,7] 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]. $^{57}$Fe Mössbauer spectroscopy and muon spin relaxation ($\mu$SR) studies of polycrystalline TbNi$_2$B$_2$C [8] confirmed the presence of a small ferromagnetic component below about 8 K previously observed via neutron diffraction [7] and magnetization measurements [10,11].

In addition to magnetic effects, strong phonon softening has been observed in superconducting $RNi_2B_2C$ 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 TbNi$_2$B$_2$C [10]. The superconducting transition temperature $T_c$ systematically decreases for $RNi_2B_2C$ (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 TbNi$_2$B$_2$C, 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 [10].

We investigated the strength of electron-phonon coupling in TbNi$_2$B$_2$C by detailed measurements of the temperature dependence of low-energy phonons in the magnetic nonsuperconducting TbNi$_2$B$_2$C 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 TbNi$_2$B$_2$C are strong and, although weaker, are of the same order of magnitude as those in superconducting $RNi_2B_2C$ compounds.
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magnetic ordering at low temperatures below 15 K with
modulated with the propagation vector
τ
the rare earth. TbNi
position of boron, scale roughly with the ionic radius of
300 K.
trons in the nearly nested regions of the Fermi surface,
ever, when phonons strongly couple to conduction elec-
tors, i.e., the lattice contants a and c as well as the z
position of boron, scale roughly with the ionic radius of
the common earth. TbNi2B2C compounds exhibit antiferro-
magnetic ordering at low temperatures below 15 K with
the magnetic Tb3+ moments aligned along the a axis and
modulated with the propagation vector \( \tau = (0.545, 0, 0) \)
\[7\] \[18\]. 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 \(^{11}\)B 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 \([\xi 0 0]\) direction, where \(\xi\) 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 nar-
row upon cooling due to decreased anharmonicity. How-
ever, when phonons strongly couple to conduction elec-
trons in the nearly nested regions of the Fermi surface,
they soften and broaden upon cooling as the Fermi sur-
face sharpens. Inelastic neutron- or x-ray-scattering mea-
surements 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 cen-
ter at \(T = 30\) K. The strongest peaks result from upward-
dispersing acoustic phonons. The inelastic peaks between
15 and 18 meV originate from downward-dispersing opt-
ical 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 excita-
tions 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 rep-
resents the background, which includes Gaussian peaks
due to CEF excitations and a straight line due to other
energy-independent sources of background. It was sub-
tracted from the data to isolate the one-phonon scatter-
ing 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 negligi-
ble. The data were divided by the Bose factor to cor-
For wave vectors between low temperatures, which is weaker but of the same or-

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 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 $(0.8,0,8)$. Our low-temperature data reveal a discrepancy in the energy range $h\omega = 8 - 10.5$ 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 or-

der of magnitude as the one found in superconducting HoNi$_2$B$_2$C. This result indicates that electron-phonon coupling in TbNi$_2$B$_2$C is strong enough 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 $T \approx 100$ K of 1 meV. Below $\approx 100$ 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 [12–13]. 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$_2$B$_2C$ [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$_2$B$_2C$ clearly demonstrate strong electron-phonon coupling in this compound. Thus we conclude that superconductivity in TbNi$_2$B$_2C$ 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 samples [1] which gives additional evidence that superconductivity in magnetic TbNi$_2$B$_2C$ 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$_2$B$_2C$ is strong enough to mediate superconductivity. This implies that its absence can only result from magnetic pair breaking.
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