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Anomalous phonon peak in the superconducting state of $\text{ErNi}_2^{11}\text{B}_2\text{C}$

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An anomalous phonon peak that appears only in the superconducting state has been reported for nonmagnetic intermetallic superconductor $R\text{Ni}_2^{11}\text{B}_2\text{C}$ ($R=\text{Y}, \text{Lu}$). In the present paper, we shall demonstrate that another rare-earth borocarbide $\text{ErNi}_2^{11}\text{B}_2\text{C}$ seems to show similar phonon anomalies. A spectral weight transfer from the above-lying TA phonon mode to the aforementioned peak was rather small, but it is consistent with the theoretical prediction for the system with smaller $2\Delta/E_{\text{ph}}$ ratio. Combining the present results with those observed in the other borocarbide systems, we suggest that such a phonon anomaly might be a common phenomenon for superconductors with a strong nesting feature of the Fermi surface.

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A family of intermetallic borocarbide superconductors $R\text{Ni}_2\text{B}_2\text{C}$ ($R=\text{Y}$ and Lu) is an attractive class of materials due to their relatively high superconducting transition temperatures T_c , (e.g. T_c up to ~ 16.5 K for $R=\text{Lu}$), and to the coexistence of the magnetic ordering with the superconducting state for compounds with magnetic rare-earth ions (e.g., Ho , Er).¹ The crystal structure of the borocarbide family at room temperature is of tetragonal $I4/mmm$ symmetry whose layered structure leads to an expectation of a possible two-dimensional character, being similar to the high- T_c cuprate superconductors.² Electronic band calculations indicate, however, that this family is composed of a three-dimensional d -band metal with a relatively high density of states of the Ni $3d$ bands at the Fermi energy E_F , and that the superconductivity is mediated by the conventional electron-phonon-type mechanism.³⁻⁵

With use of the neutron scattering technique, we previously reported that $\text{YNi}_2^{11}\text{B}_2\text{C}$ ($T_c=14.2$ K) gives rise to an anomalous sharp peak at ~ 4 meV in its phonon spectra in the superconducting state.⁶⁻⁹ This peak is gradually suppressed by applying a magnetic field, and vanishes above H_{c2} . From its response to the external field and temperature, we concluded that this peak is intimately associated with its superconducting state. The peak energy ~ 4.0 meV is very close to a BCS superconducting gap $2\Delta = 3.5k_B T_c \sim 4.3$ meV, and the anomaly is located at a wave vector that corresponds to the nesting vector of the Fermi surface of this compound. In addition, by a survey of equivalent positions in the Q space, we identified this peak to be of phonon origin with polarization parallel to the $[001]$ axis and with a propagation vector $[100]$. Furthermore, we demonstrated that this peak grows by absorbing the spectral weight of the above-lying transverse acoustic phonon mode, and that the sum of the intensity of two peaks are conserved (the sum rule of the

phonon intensity). These results strongly indicate that the anomalous peak is related to the nesting of the Fermi surface and to the above-lying transverse acoustic phonon mode. Following our work, a similar phonon anomaly was also (re-) confirmed in the superconducting state of $\text{LuNi}_2\text{B}_2\text{C}$ and $\text{YNi}_2\text{B}_2\text{C}$.¹⁰⁻¹²

Allen *et al.*¹³ extended the calculation of the phonon self-energy effect to the region of a finite q , and obtained the phonon resonance peak at $E=2\Delta$ and the sum rule of the spectral weight between the resonance peak and above-lying phonon peak. This theory, however, does not incorporate the nesting feature of the Fermi surface.

Kee and Varma, on the other hand, proposed a different mechanism to explain the anomalous phonon peak.¹⁴ They calculated an electronic polarizability near extremum vectors of the Fermi surface and showed that this phonon peak may appear in the superconducting state with energy just below the superconducting gap 2Δ . Their theory also demonstrated that a phonon spectral weight is transferred from the above-lying phonon to the resonance peak, being consistent with our experimental results and interpretations. We should note here that there are, however, some discrepancies between the theoretical predictions and the experimental results. In the theory, the energy of the anomalous peak cannot exceed its superconducting gap energy 2Δ so that the temperature dependence of the energy position of this peak should follow a BCS-like curve. The temperature dependence of 2Δ observed by tunneling spectroscopy shows a BCS-like curve.¹⁵ On the other hand, the temperature dependence of the energy position of this peak observed by neutron scattering shows almost temperature-independent behavior.

In order to understand the phonon anomaly in the superconducting state, it is very important to further accumulate reliable experimental results, and examine the consistency

between the theory and observations. For instance, the latter theory predicts that a transfer of the spectral weight from the above-lying phonon to the anomalous peak becomes smaller, when the $2\Delta/E_{\text{ph}}$ becomes smaller where E_{ph} is the energy of the above-lying phonon peak. Therefore, it is very useful to perform an experimental test of such a theoretical prediction by employing another system with a different ratio of $2\Delta/E_{\text{ph}}$. In the following, we shall report inelastic neutron scattering results on the $\text{ErNi}_2^{11}\text{B}_2\text{C}$ system. The superconducting gap energy 2Δ of our $\text{ErNi}_2^{11}\text{B}_2\text{C}$ sample ($T_c = 8.6$ K) is ~ 2.6 meV, which can be estimated as $2\Delta = 3.5k_B T_c$ from the BCS theory for a weakly coupled superconductor. We found that the inelastic intensity at around ~ 3 meV shows a small increase in the superconducting state. The spectral weight transfer of the cross section is unclear but is fully consistent with the theoretical prediction for the system with a small $2\Delta/E_{\text{ph}}$ value as explained below.

Note that $\text{ErNi}_2^{11}\text{B}_2\text{C}$ undergoes a spin density wave (SDW) transition at ~ 6 K, whose magnetic structure was determined to be a transversely polarized planar sinusoidal type with a propagating wave vector $q = 0.553a^*$ by neutron diffraction measurements.^{16,17} The magnetic transition at T_N is accompanied with a structural tetragonal-to-orthorhombic transition.¹⁸ Furthermore, the coexistence state between weak ferromagnetism (WFM) and superconductivity below $T_{\text{WFM}} \sim 2.3$ K was suggested by magnetization and specific measurements,¹⁹ and was microscopically confirmed by utilizing polarized and unpolarized neutron scattering techniques.^{20–22} In a previous paper, we determined a precise magnetic structure in the WFM state, where one from twenty Er moments contributes the WFM order.²³ We would like to stress here that $\text{ErNi}_2\text{B}_2\text{C}$ is the first material in which a stable coexistence of the WFM with superconductivity was microscopically confirmed.

For the present neutron scattering experiment, we used large single crystals of $\text{ErNi}_2^{11}\text{B}_2\text{C}$ (total volume of ~ 1 cm³). The single crystals were grown by a floating zone method, and a detailed procedure was previously reported by Takeya *et al.*²⁴ The superconducting transition temperature of our sample was determined to be 8.6 K (midpoint) with a transition width of 0.4 K by magnetization measurements. Note that T_c of our sample is about 2 K lower than the one obtained by the flux method. Very recently it was found that the annealing process raises T_c up to 10.5 K but causes no change for T_N and T_{WFM} . We interpreted that metallurgical microstructures in the crystal decrease T_c . The results in the present paper were taken with the nonannealed sample ($T_c = 8.6$ K).

Inelastic neutron scattering measurements were performed with the triple-axis spectrometer GPTAS (4G) installed in the JRR-3M research reactor at Japan Atomic Energy Research Institute, Tokai, Japan. The spectrometer was operated in a constant k_f mode with a final neutron momentum of $k_f = 3.825 \text{ \AA}^{-1}$. The (002) reflection of pyrolytic graphite (PG) was utilized as monochromating and analyzing the neutron energy. A PG filter was placed to reduce higher-order contaminations. A combination of collimators of 20'-40'-40'-40' or 40'-80'-80'-80' was selected at standard positions from inpile to detector. The sample was oriented

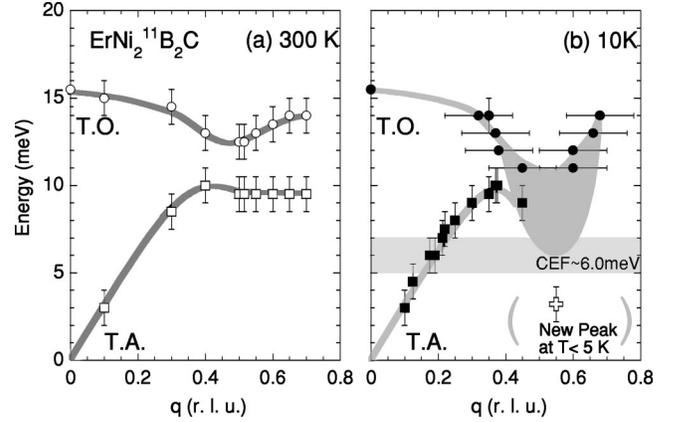


FIG. 1. Dispersion curves for phonon excitations at 300 K (left) and 10 K (right), respectively. The energy position of the anomalous peak, which is observed only below T_c , is indicated by a thick cross symbol in the right panel. For more details, see text. Solid curves are guides to the eye.

with their [010] axis vertical to the scattering plane, and it was set in an aluminum can with exchange He gas, and was mounted to a liquid-He-type cryostat. The sample temperature ranging from 1.45 K to 300 K was controlled within an accuracy of 0.1 K. In the present study, we measured scattering profiles along $Q = (q, 0, 8)$, $(1 - q, 0, 7)$, and $(1 - q, 0, 9)$ by constant- Q and constant- E scans with neutron energy transfer $\Delta E < 20$ meV. In this energy range and Q positions, one can observe the transverse acoustic and lowest-lying optical phonon peaks that belong to the same Δ_4 symmetry.

The dispersion curves for these two phonon branches at 300 K and 10 K are summarized in Fig. 1. For $q < 0.4$, two branches show little temperature dependence. By contrast, the first optical branch for $q > 0.4$ shows a large softening of the energy and a broadening of the width. Because of these, it is difficult to determine the energy positions of these two branches separately at low temperatures, and such a region is indicated by a hatch in Fig. 1(b). The softening is strongest at around $q \sim 0.55$ which corresponds to the nesting vector of the Fermi surface of this compound.²⁵ Therefore, the softening should be attributed to the Kohn anomaly as was done in the previous studies. In addition, a crystal field excitation peak was clearly observed at $E = 6.0$ meV at low temperature.²⁶ With further lowering temperature below T_c , we noticed a very weak increase of intensity at around $(0.55, 0, 8)$ at the energy transfer of ~ 3 meV, which is marked by an open thick cross symbol in Fig. 1(b).

Figure 2 shows the temperature dependence of phonon profiles at selected temperatures observed at $(0.548, 0, 8)$ and $(0.452, 0, 9)$ with an energy transfer of ~ 2 meV $< \Delta E < 20$ meV. At 300 K, the profile at $(0.548, 0, 8)$ shows a large peak at 13 meV with a shoulderlike feature at 9.5 meV, while that at $(0.452, 0, 9)$ consists of one peak at 9.5 meV. The difference of the profiles is due to the difference of the cross section at these two Q positions. By comparing two profiles, one can easily determine positions of two respective peaks. The peak at higher energy is identified as the first optical phonon mode. This optical phonon peak shows a drastic soft-

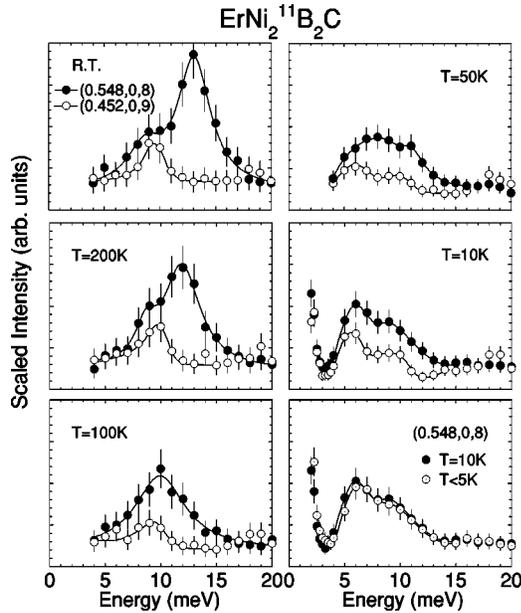


FIG. 2. Phonon profiles of a constant- Q scan observed at $Q = (0.548, 0.8)$ and $(0.452, 0.9)$ in $\text{ErNi}_2^{11}\text{B}_2\text{C}$. Curves are guides to the eye. By taking account of systematic errors in inelastic neutron scattering experiments, we adopt 3σ as a size of error bars.

ening with decreasing temperature, and it merges into the acoustic mode below 100 K. In addition, with further decreasing temperature, an additional peak due to the crystal field excitation emerges at $E=6.0$ meV. At 10 K, a very broad peak at ~ 9.0 meV and a rather sharp peak at ~ 6.0 meV were observed. Furthermore, by careful examination of the phonon profiles in a right-bottom panel, we recognized a subtle increase of the scattering intensity at around 3 meV below T_c . The energy position of the change ~ 3 meV is very close to the superconducting gap energy ~ 2.6 meV calculated for this material, indicating a similar phonon anomaly occurs in $\text{ErNi}_2^{11}\text{B}_2\text{C}$.

As mentioned above, however, the system shows a structural phase transition at $T_N \sim 6$ K, which causes a change of the crystal symmetry from tetragonal to orthorhombic (a change to the lower symmetry).¹⁸ Therefore one can expect that the lift of the degeneracy can give rise to a new phonon branch at $E \sim 3$ meV. In order to check whether or not the increase of the intensity at 3 meV is related to the structural transition at T_N , we measured the temperature dependence of the peak intensity at $Q = (0.548, 0.8)$ and $E = 3.25$ meV, and the result is shown in Fig. 3. The result indicates that the intensity increases at T_c but not at T_N .

Furthermore, although a spectral weight transfer from the above lying TA phonon peak to the anomalous peak is clearly observed in the $\text{YNi}_2^{11}\text{B}_2\text{C}$ and $\text{LuNi}_2^{11}\text{B}_2\text{C}$ cases,

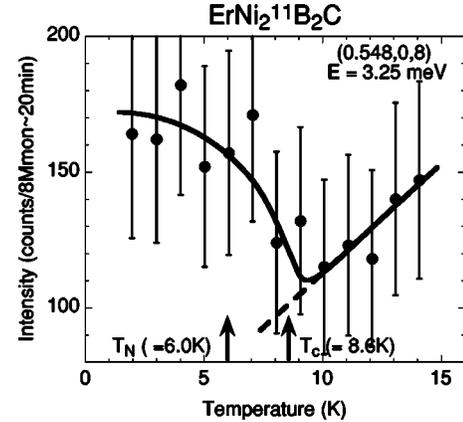


FIG. 3. Temperature dependence of scattering intensity at $Q = (0.548, 0.8)$ and with $E = 3.25$ meV. The curve is drawn for a guide to the eye. By taking account of systematic errors in inelastic neutron scattering experiments, we adopt 3σ as a size of error bars.

it is not so clear in the present case and the intensity of the anomalous peak is relatively small. The aforementioned theory suggests, however, that the spectral weight transfer is smaller in the system with a smaller $2\Delta/E_{\text{ph}}$ value.¹⁴ The ratio $2\Delta/E_{\text{ph}}$ is ~ 0.6 for $\text{YNi}_2^{11}\text{B}_2\text{C}$ and ~ 0.3 for $\text{ErNi}_2^{11}\text{B}_2\text{C}$. Therefore, the present result is not inconsistent with this prediction.

With these considerations, we conclude that the change of inelastic intensity at ~ 3 meV is attributed to the same phonon anomalies which observed in the Y and Lu borocarbides.⁶⁻¹² In conclusion, inelastic neutron scattering measurements on ^{11}B -substituted $\text{ErNi}_2^{11}\text{B}_2\text{C}$ were performed with large single crystals. Below T_c , a change of inelastic intensity with a phonon character was observed at around $Q = (0.55, 0.8)$ and with $E \sim 3.0$ meV. The energy is very close to the its superconducting gap $2\Delta \sim 2.6$ meV of $\text{ErNi}_2^{11}\text{B}_2\text{C}$. Although the transfer of the cross section from the above-lying phonon mode to the anomalous peak is less distinct in the Er compound, it is consistent with theoretical predictions for phonon anomalies for the system with a small $2\Delta/E_{\text{ph}}$ value. The present results indicate that the change in $\text{ErNi}_2^{11}\text{B}_2\text{C}$ is again related to its superconducting state, and the accumulating results establish that the anomalous peak is a common feature in the superconducting borocarbide systems.

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