Anomalous Phonon Scattering Below T_c in $YNi_2{}^{11}B_2C$

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Inelastic neutron scattering measurements were performed on large volume (~ 2 cm^3) single crystals of ^{11}B substituted intermetallic superconductor $YNi_2{}^{11}B_2C$. With decreasing temperature, a gradual softening of a phonon spectrum is observed at around $Q = (0.525, 0, 8)$, similar to the case of $LuNi_2B_2C$ reported by Dervenagas *et al.* [Phys. Rev. B **52**, R9839 (1995)]. In addition, we discovered that an intense new peak appears below $T_c = 14.2$ K. With application of magnetic field, however, it is easily suppressed and disappears by $H = H_{c2}$ (~ 4.7 T at $T = 5.5$ K). These results strongly indicate that this new peak is undoubtedly associated with its superconducting state. [S0031-9007(96)01653-5]

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The newly discovered borocarbide intermetallic superconductors $RETM_2B_2C$ (RE = Y and Lanthanoid, TM = Ni, Pd, Pt) [1] exhibit a number of attracting properties for a study of superconductivity due to their relatively high superconducting transition temperatures T_c and due to the competition between superconductivity and magnetism [2,3]. In contrast to the high T_c cuprate superconductors, recent intensive studies on the borocarbide superconductors have revealed that the superconductivity of this family is three dimensional rather than two dimensional despite a RE-C and Ni_2-B_2 layered structure [4]. Band calculations suggest their superconductivity is a conventional BCS superconductivity mediated by the electron-phonon couplings [5]. A relatively small H_{c2} activated intensive experimental studies on voltex lattice [6].

Despite these attractive features of this family, there are some drawbacks for a neutron inelastic scattering study. Most of the measurements on $RETM_2B_2C$ so far have been performed on the arc-melted polycrystalline samples due to the difficulty of growing a large single crystal. Although some groups have succeeded in making single crystals by flux method, those are a few mm in linear dimension. Dervenagas *et al.* have successfully performed inelastic neutron scattering measurements by aligning two $LuNi_2B_2C$ single crystals of $5 \times 5 \times 0.5$ mm^3 in size, and revealed that phonon branches measured along the [100] direction from (008) exhibit continuous softening near the zone-boundary point G_1 from room temperature [7]. At around the same wave vector, calculations on electron susceptibility indicate that the electron susceptibilities in these compounds show a local maximum [8]. It is also reported that the compounds with magnetic rare-earth ions such as $HoNi_2B_2C$ [9,10] and $ErNi_2B_2C$ [11,12] exhibit incommensurate magnetic phases with similar incommensurate vector q_a along the [100] direction, i.e., $q_a = 0.585a^*$ for $HoNi_2B_2C$ and $q_a = 0.553a^*$ for $ErNi_2B_2C$, respectively. This behavior reflects a feature of strong nesting of the Fermi surface in these compounds. Accordingly, the softening of phonon spectra observed in $LuNi_2B_2C$ was interpreted as a Kohn anomaly.

Recently one of us (H. T.) succeeded in growing a large single crystal of a family of $RETM_2B_2C$ by a floating zone method which has a sufficiently large size for neutron inelastic measurements, and this success enabled us to perform a systematic study of inelastic responses in a family of $RETM_2B_2C$. As a first step, we have performed an intensive study of inelastic responses in $YNi_2{}^{11}B_2C$. We have confirmed that a gradual phonon softening also occurs in $YNi_2{}^{11}B_2C$ below room temperature. Surprisingly, however, we have discovered that a sharp new peak grows below T_c in a narrow Q region at around $Q = (0.525, 0, 8)$ and $E \cong 4.0$ meV. Applying magnetic field, this new peak disappears by H_{c2}. Although the origin of the peak is unclear at the moment, this peak is conclusively attributed to its superconducting state.

To avoid strong absorption due to ^{10}B, ^{11}B-isotope-enriched (99 at.% up) large single crystals $YNi_2{}^{11}B_2C$ of 7 mm$^{\phi} \times 100$ mm in size were grown for the present neutron scattering experiments. Detailed procedure of the crystal growth was reported by Takeya *et al.* [13]. The orientation of the crystal was determined by a Laue method, and the sample ingot was cut into pieces of 30 mm in length. The superconducting transition temperature of the obtained single crystals was determined to be 14.2 K (midpoint) with the transition width of 0.5 K by magnetization measurements.

Inelastic neutron scattering measurements were performed with the triple axis spectrometer GPTAS (4G) installed in the JRR-3M at JAERI, Tokai. The spectrometer was operated in a constant k_f mode with $k_f = 3.825$ Å^{-1}, and pyrolytic graphite crystals were utilized as monochromator and analyzer. Collimators of $40'$-$40'$-$40'$-$40'$ or $20'$-$20'$-$20'$-$20'$ were placed at standard positions

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from inpile to detector. $YNi_2{}^{11}B_2C$ has a body centered tetragonal structure (space group I4/mmm), consisting of alternative stacking of RE-C and Ni_2-B_2 layers along the c axis [14]. Three single crystals were oriented within ~0.1 degree with their [010] axes vertical to the scattering plane, and phonons which propagate to the [100] direction with polarization vector [001] were measured in the $(h, 0, l)$ zone. For the magnetic field experiments, a conventional superconducting magnet was utilized to produce a vertical field of $0 < H \leq 6.0$ T perpendicular to the scattering plane and parallel to the [010] axis. The sample temperature ranging from 1.6 to 350 K was controlled within accuracy of 0.1 degree.

The temperature dependences of the phonon spectra observed at $Q = (0.5, 0, 8)$ ($T > 15.2$ K) and $Q = (0.55, 0, 8)$ ($T < 15.2$ K) are shown in Fig. 1. A gradual softening was observed from room temperature to $T \sim 30$ K (upper panel), as was reported in Ref. [7] for the Lu compound, in which this softening was interpreted as a Kohn anomaly due to strong nesting on the Fermi surface. However, the peak position shows no shift below ~30 K, and the profiles at 30 and 15.2 K are essentially identical. As shown in the lower panel, on the other hand, we found that a new peak emerges at ~4.75 meV with further decreasing temperature below T_c. Note that there is considerable intensity at ~7 meV even at 7.5 K, indicating that the spectrum consists of two components in the superconducting phase. We also note that the Lu compound the acoustic and optical branches at $Q \sim (0.5, 0.8)$ yield peaks at ~4 meV and ~9 meV at 10 K, whereas in the Y compound, we observed that the optical branch yields a peak at ~14 meV in the superconducting phase.

To further examine the behavior of two components in the profile, the temperature dependence of each component was observed at $Q = (0.525, 0, 8)$, $E = 4.0$, and 7.0 meV, respectively. As shown in Fig. 2, the intensity of the new peak at 4.0 meV shows a clear onset at T_c, and its temperature dependence is akin to the order parameter of superconductivity. In contrast, the spectral weight of the soft phonon component observed at 7 meV shows a slight decrease below T_c, but substantial intensity remains down to ~2.5 K, supporting the two-peak structure of the spectral shape. One might suspect that the new peak grows by absorbing the spectral weight from the above-lying soft phonon mode, and we shall demonstrate that it is indeed the case because the sum of integrated intensity of the new peak and the soft phonon is conserved as shown later.

If this new component truly reflects the superconducting state in the $YNi_2{}^{11}B_2C$ sample, the magnetic field can suppress it above H_{c1} and should wipe it out above H_{c2}. Two critical fields in our samples are determined to be $H_{c1} \sim 0.018$ T and $H_{c2} \sim 4.7$ T at 5.5 K, respectively. The field dependence of the phonon spectra observed at $Q = (0.55, 0, 8)$ is shown in Fig. 3. With application of magnetic field, the intense peak at $E = 4.75$ meV broadens progressively, and the spectrum at $H = 6.0$ T recovers the profile observed in the normal state at 15.2 K (See Fig. 1). Note that at 2.0 T, one can clearly recognize two peaks in the profile, further supporting our identification of the two-peak structure of the spectral shape. We also measured the field dependence of the peak intensity at $Q = (0.55, 0, 8)$ with $E = 4.75$ meV. The result was shown in Fig. 4. The peak intensity shows a continuous decrease with increasing magnetic field and completely

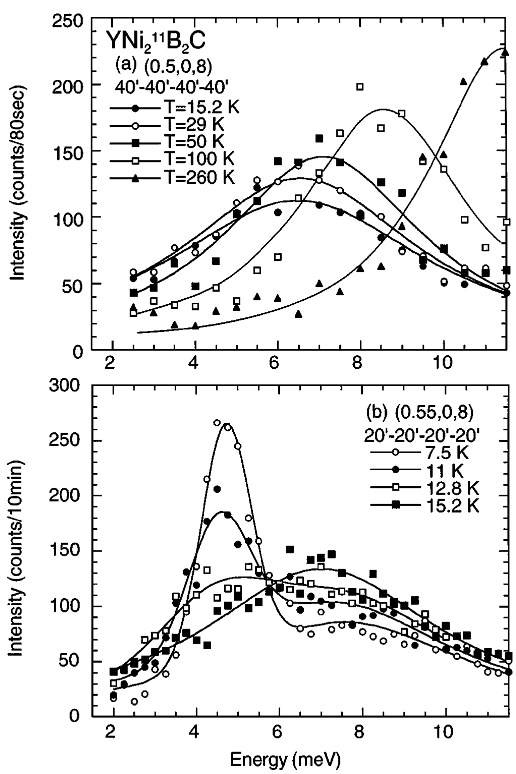

FIG. 1. Temperature dependence of phonon profiles. Upper panel: $T > T_c$ and $Q = (0.5, 0, 8)$. Lower panel: $T \lesssim T_c$ and $Q = (0.55, 0, 8)$.

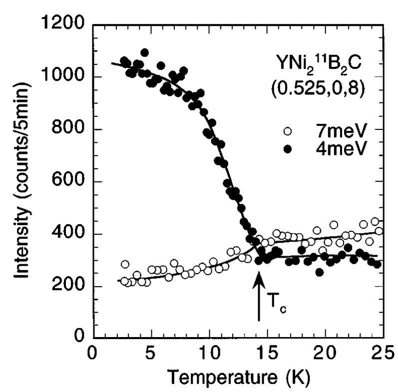

FIG. 2. Temperature dependence of inelastic scattering intensity at (0.525, 0, 8). Filled and open symbols indicate the intensity at $E = 4.0$ and 7.0 meV, respectively.

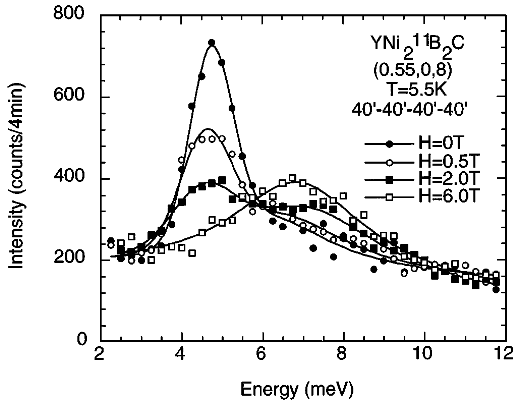

FIG. 3. Field dependence of the profiles measured at $Q = (0.55, 0, 8)$. Solid lines are guides for the eye.

disappears by H_{c2}. We conclude from these observations that the new peak should be attributed to the superconducting state in $YNi_2{}^{11}B_2C$.

From constant-Q and constant-E scans, we mapped out the distribution of the new peak, and established that the new peak appears in a narrow Q region of $Q = (q, 0, 8)$ with $0.4 \lesssim q \lesssim 0.7$. It is almost resolution limited in energy, and exhibits a weak dispersion with the range of $4.0 < E < 7.0$ meV. By a survey of equivalent positions in Q space, we identified that this peak has a phonon origin with polarization vector parallel to the [001] axis and with a propagation vector [100].

To further check the nature of the new peak, we examined the integrated intensities of the new peak and of the above-lying soft phonon mode as a function of q, T, and H, after correcting the thermal occupation factor. The upper panel (a) in Fig. 5 shows the q dependence of the two components at $T = 1.6$ and 15.2 K for $0.475 \leq q \leq 0.675$. One can see that the data points of the sum of the two components at 1.6 K (filled triangles) come very close to those of the integrated intensity of the soft phonon at 15.2 K in the normal state (open triangles). The lower panels (b) and (c) show the T and H dependences of the two components observed at $q = 0.55$, respectively. Clearly the sum of the integrated intensity of the two components is conserved throughout the variation of T or H, indicating that the new peak is intimately related with the above-lying soft phonon mode.

We also found that the energy of the new peak is very close to the superconducting gap which can be estimated from BCS theory as $2\Delta = 3.5k_BT_c$ ($\sim$4.3 meV for $T_c = 14.2$ K) for a weakly-coupled superconductor. To examine the relation of the BCS gap and the energy of the new peak, we have measured the T dependence of the peak position of the new peak at $Q = (0.525, 0, 8)$ (not shown). We found, however, that the observed T dependence of the peak position is weaker than that of the BCS gap. Other intriguing experimental findings are that the new peak is strongest at the position which corresponds to the nesting vector of the Fermi surface, and that the position of the new peak coincides with that of

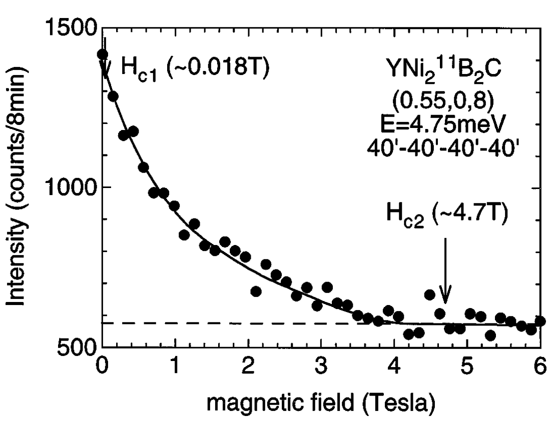

FIG. 4. Field dependence of the peak intensity observed at $Q = (0.55, 0, 8)$ and $E = 4.75$ meV. The arrows indicate the critical magnetic field H_{c1} and H_{c2} determined by magnetization measurements. A solid curve is a guide for the eye.

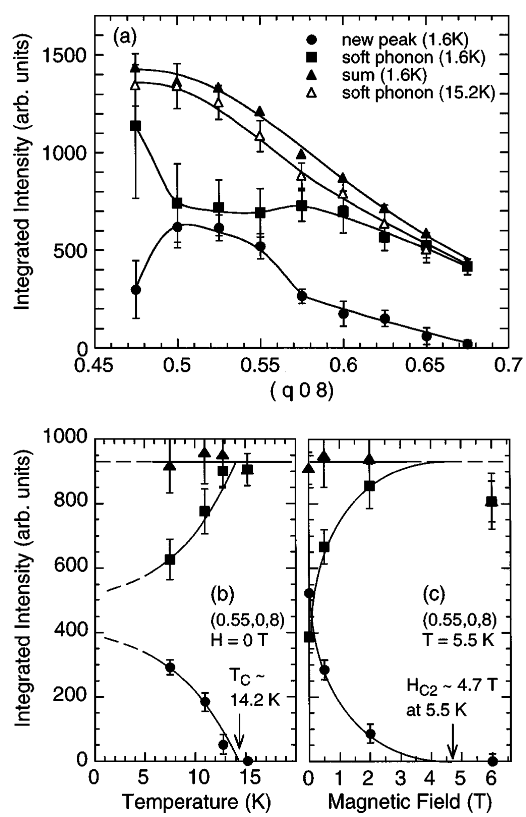

FIG. 5. Panels (a), (b), and (c) show q, T, and H dependences of the integrated intensity of the new peak and of the soft phonon, respectively. The data were analyzed after the occupation factor correction. The circle, square, and triangle symbols indicate the integrated intensity of the new peak, the soft phonon, and the sum of two peaks, respectively. The curves are drawn for guides to the eye. For the field dependence, the data at 6.0 T were recorded with a slightly different spectrometer setting.

the Kohn-type phonon anomaly observed above T_c. We believe that these coincidences cannot be accidental, and that these factors are essential to elucidate the mechanism of the new excitation in $YNi_2{}^{11}B_2C$ reported here.

The phonon frequency and the lifetime are expected to be strongly influenced by superconductivity through the phonon self-energy effects [15]. In the weak-coupling case, one expects a narrowing of the width and a decrease of the peak frequency for the phonons with frequency lower than the superconducting gap energy, while an increase of the peak frequency for those with higher frequency. One of well-known results on the phonon softening in the conventional superconductors is an inelastic neutron study on Nb_3Sn [16]. For one of High T_c cuprate superconductors $YBa_2Cu_3O_7$, similar anomalies in phonon spectra have been also reported recently [17]. Compared with these results, however, we find that the two-peak structure of the phonon spectrum in the $YNi_2{}^{11}B_2C$ sample is qualitatively different.

On the other hand, we learned that a similar peak was observed by Raman scattering measurements in a charge density wave (CDW) superconductor, $2H$-$NbSe_2$ [18]. This material shows a CDW transition at $T_d \sim 33$ K and becomes superconducting below $T_c \sim 7.2$ K. In the superconducting phase, the growth of a new sharp peak was observed close in energy to the BCS gap 2Δ (~ 17 cm^{-1}) and below the CDW-induced phonon mode (~ 40 cm^{-1}). By applying magnetic field, the new peak was suppressed, and the spectral weight was transferred to the CDW-induced phonon mode. It was claimed that the 2Δ gap mode borrowed its Raman activity from the CDW via electron-phonon coupling.

Littlewood and Varma (LV) developed the gauge invariant theory on this phenomenon [19]. Their theory predicts that the superconducting gap yields the two collective modes, the gap amplitude mode and the gap phase mode, but only the former is relevant under the Coulomb interaction. Unfortunately, the gap mode is not experimentally observable in most superconductors because of a lack of suitable interactions with photons or with neutrons. In $2H$-$NbSe_2$, however, the CDW-induced phonon mode and superconducting gap are coupled through the conduction electron density of states at the Fermi surface, as was demonstrated by the pressure effects on the phase transition temperatures T_c and T_d [20]. LV suggested that, having an appropriate interaction, the new peak in $2H$-$NbSe_2$ can be explained as a coupled excitation of a gap-amplitude mode and the CDW-induced phonon mode. In the superconducting phase, the gap-amplitude mode gives rise to a sharp feature in the dynamical scattering function $S(\omega)$ of the CDW phonon at energy close to 2Δ.

Although there is a close similarity between the present new peak in $YNi_2{}^{11}B_2C$ and the 2Δ excitation in $2H$-$NbSe_2$, the new peak in $YNi_2{}^{11}B_2C$ appears at around the G_1 point instead of the Brillouin zone center, and $YNi_2{}^{11}B_2C$ shows no CDW transition. Nevertheless, we feel that aforementioned theory can be a good candidate for the present case provided that the Kohn anomaly-induced soft mode can play an identical role with the CDW-induced phonon mode in the case of $2H$-$NbSe_2$.

In conclusion, inelastic neutron scattering measurements on ^{11}B substituted $YNi_2{}^{11}B_2C$ were performed with high quality large single crystals. Below T_c, a new sharp excitation with phonon character appears at around $Q = (0.525, 0, 8)$ and $E \cong 4.0$ meV. The temperature dependence of the new peak is similar to the superconducting order parameter, and the peak grows by absorbing the spectral weight of the above-lying soft phonon mode. This new peak is gradually suppressed by magnetic field and vanishes above H_{c2}. These results establish that the new peak is related with the superconducting state and with the above-lying soft phonon mode in $YNi_2{}^{11}B_2C$.

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