## Thermal conductivity of $RNi_2B_2C$ (R = Y,Ho) single crystals

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We have studied the thermal conductivity  $\kappa$  of high-quality single crystals of YNi<sub>2</sub>B<sub>2</sub>C and HoNi<sub>2</sub>B<sub>2</sub>C. In both compounds,  $\kappa$  in the normal state at low temperatures is dominated by electrons. In YNi<sub>2</sub>B<sub>2</sub>C,  $\kappa$  shows a pronounced enhancement around 4 K well below the superconducting transition temperature  $T_c$ =15.6 K. It may be due to the increase of the phonon contribution below  $T_c$  as a result of the decrease of the number of normal electrons as scattering centers of phonons. This enhancement is suppressed largely by applying the magnetic field of  $H_{c1}$  because phonons are scattered by vortices. In HoNi<sub>2</sub>B<sub>2</sub>C,  $\kappa$  shows a discontinuous increase at the Néel temperature  $T_N$ =4.6 K and a clear decrease below  $T_c$  ~3.6 K. [S0163-1829(96)03130-X]

Since the recent discovery of the superconductivity in  $RNi_2B_2C$  (R=Y, Ho, etc.) (Refs. 1,2), extensive studies have been performed in the related compounds which have relatively high superconducting transition temperature  $T_c$ . Their crystal structure is of the layered type and they have attracted much attention in relation to high- $T_c$  cuprates. However, the band calculation indicates that the electronic structure is three dimensional and many experimental results suggest that these materials are the conventional phonon mediated s-wave superconductor.<sup>3,4</sup> However, these materials also exhibit unexpected behavior from an s-wave superconductor. For example, in YNi<sub>2</sub>B<sub>2</sub>C and LuNi<sub>2</sub>B<sub>2</sub>C, the coherent peak in the NMR relaxation rate below  $T_c$  has not been observed (Ref. 5) and the specific heat shows  $\sim T^3$  behavior in a wide range of temperature below  $T_c$ . 6,7 Further studies are necessary to clarify the mechanism of the superconductivity in the present system. The thermal conductivity  $\kappa$  is very useful to investigate the nature of the superconducting state and has been studied extensively on heavy Fermion superconductors and high- $T_c$  cuprates in recent years. In high- $T_c$  cuprates, it is well known that the pronounced enhancement of  $\kappa$  is observed below  $T_c$  and the recent results under the magnetic field indicate that its origin is the huge enhancement of the lifetime of quasiparticles below  $T_c$ . In a conventional low- $T_c$  s-wave superconductor, the phonon contribution to  $\kappa$ increases below  $T_c$  due to the decrease of the number of normal electrons as scattering centers of phonons and the electron contribution decreases rapidly below  $T_c$ . Whether the observed thermal conductivity increases or decreases below  $T_c$  depends on which contribution of phonons and electrons is dominant.

The thermal conductivity of  $R\mathrm{Ni_2B_2C}$  has not yet been reported. In order to get information on the electron-phonon interaction and the low-energy excitation in the superconducting state in the present system, the thermal conductivities of high-quality single crystals of  $Y\mathrm{Ni_2B_2C}$  samples 1 and 2 and  $Ho\mathrm{Ni_2B_2C}$  were measured. All the samples used in the

present experiments are single crystals grown by the floating-zone method which is described elsewhere. The samples were in the form of a parallelepiped with dimensions  $0.7 \times 0.7 \times 4 \sim 7$  mm<sup>3</sup>. The thermal conductivity  $\kappa$  was measured by the usual steady-state method. The temperature gradient of both ends of the samples in the zero magnetic field at high temperatures and under the magnetic field was measured by the AuFe-Chromel thermocouple and two Cernox thermometers, respectively. The electrical resistivity  $\rho$ was measured by the usual four-probe method. The error bar of the absolute value of  $\kappa$  and  $\rho$  is about 10–15 % and 20– 30 %, respectively, depending on the sample size. The residual resistivity of YNi<sub>2</sub>B<sub>2</sub>C samples 1 and 2 and HoNi<sub>2</sub>B<sub>2</sub>C are  $\sim 2.0$ , 4.1, and 3.1  $\mu\Omega$  cm, respectively. The de Haasvan Alphen effect was observed in the samples of YNi<sub>2</sub>B<sub>2</sub>C sample 1 cut from the same ingot. 10 The magnetostriction was measured by the three terminal capacitance method.<sup>11</sup> The specific heat was measured by the usual adiabatic heat pulse method for the same HoNi<sub>2</sub>B<sub>2</sub>C crystals as those used in the  $\kappa$  and  $\rho$  with a weight of 56 mg.

Figure 1 shows the temperature dependence of the thermal conductivity  $\kappa$  of YNi<sub>2</sub>B<sub>2</sub>C sample 1. The inset shows the temperature dependence of the reduced Lorenz number  $L/L_0$  of the same sample which was derived from the observed  $\kappa$  and  $\rho$ . Here,  $L_0 = 24.5 \text{ nW } \Omega \text{ K}^{-2}$  is the Sonmerfeld value and  $L = \kappa \rho / T$ . Along the a axis,  $L/L_0$  is nearly 1 at low temperatures below  $\sim 20$  K in the normal state. The absolute values of  $\kappa$  of YNi<sub>2</sub>B<sub>2</sub>C sample 2 at T=20 K are 110 and 103 mW/K cm along the a and c axes, respectively. The difference of the absolute value of  $\kappa$  at low temperatures between samples 1 and 2 by a factor of two is almost the same as that of the residual resistivities between these samples. These results indicate that in the normal state at low temperatures,  $\kappa$  is limited by the impurity scattering, i.e., the elastic scattering of electrons by impurities is dominant and heat current is carried mainly by electrons. Rough estimation of the electronic thermal conductivity  $\kappa_e$  at low temperature

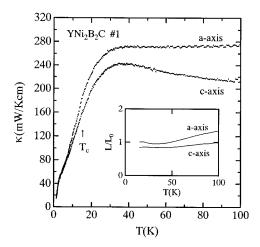


FIG. 1. Temperature dependence of the thermal conductivity with heat current J parallel to the a and c axes of YNi<sub>2</sub>B<sub>2</sub>C, sample 1. The inset shows the temperature dependence of the reduced Lorenz number  $L/L_0$  of sample 1. See the text in detail.

by using the results of the electronic specific heat (Ref. 6) and the electrical resistivity is consistent with the observed result.  $\kappa$  shows a small but clear decrease at  $T_c = 15.6$  K and a pronounced enhancement around 4 K which is clearly seen in an expanded scale in Fig. 2. This characteristic behavior is observed also in sample 2 with twice larger residual resistivity. At high temperatures above  $\sim 40$  K,  $\kappa$  shows the anisotropic behavior, i.e.,  $\kappa$  with heat current J parallel to the a axis is larger than that parallel to the a axis. This anisotropic behavior of  $\kappa$  at high temperatures is common to YNi<sub>2</sub>B<sub>2</sub>C samples 1 and 2 and HoNi<sub>2</sub>B<sub>2</sub>C, while their electrical resistivities are almost isotropic.  $\kappa$  parallel to the a axis of

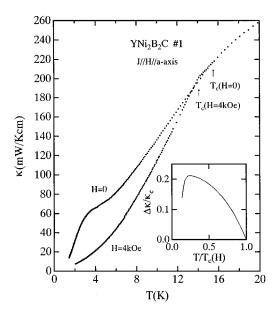


FIG. 2. Temperature dependence of the thermal conductivity with heat current J parallel to the a and c axes of  $YNi_2B_2C$  sample 1 under the longitudinal magnetic field. The inset shows the temperature dependence of the difference of the temperature dependence of the thermal conductivities  $\kappa_c$  normalized at  $T_c(H)$  between under  $H\!=\!0$  and  $H\!=\!4$  kOe.

YNi<sub>2</sub>B<sub>2</sub>C sample 2 does not show a peak as is seen in sample 1 which may be due to a large residual resistivity in this sample. With the increase of temperature,  $L/L_0$  once becomes smaller than 1, again increases and becomes larger than 1 above  $\sim$ 50 K. The fact that  $L/L_0 > 1$  at high temperatures above ~50 K indicates an increase of the phonon contribution to  $\kappa$ . The fact that  $L/L_0 < 1$  in the intermediatetemperature region indicates the existence of the inelastic scattering for electrons. While the effect of the scattering of electrons by long-wavelength acoustic phonons on  $\rho$  is small, the thermal conductivity is largely affected through the vertical transition. Different from  $\kappa$  parallel to the a axis, in the case parallel to the c axis,  $L/L_0$  is smaller than 1 in a wide range of temperature. This situation is common to all the samples studied here, which reflects the anisotropic behavior of  $\kappa$  as mentioned above. Further studies are necessary to understand this anisotropy.

Figure 2 shows the temperature dependence of  $\kappa$  parallel to the a axis in the zero magnetic field and under the longitudinal magnetic field of 4 kOe. The thermal conductivity in the zero magnetic field shows the characteristic behavior, i.e., an anomaly at  $T_c$  and a pronounced maximum at  $\sim$ 4 K as mentioned above. On the other hand, the thermal conductivity under the magnetic field of 4 kOe shows a rapid decrease below  $T_c$  (H=4 kOe), which is common to conventional superconductors in which heat current is carried mainly by electrons. Figures 3(a) and 3(b) show the longitudinal magnetic-field dependence of the thermal conductivity parallel to the a axis of  $YNi_2B_2C$  sample 1. The  $\kappa$  value is largely suppressed by applying a small magnetic field corresponding to  $H_{c1}$  and, with further increase of the magnetic field, it is restored to the normal-state value above  $H_{c2}$ . The hysteresis is observed below  $H_{c1}$ , which is shown in the result at T = 4.3 K. The similar results are observed also in  $\kappa$ parallel to the a axis under the transverse magnetic field parallel to the c axis. The rapid suppression of the thermal conductivity at  $H>H_{c1}$  indicates that heat current is suppressed largely by the existence of vortices. The inset in Fig. 2 shows the temperature dependence of the difference of the thermal conductivities normalized at  $T_c(H)$  between under H=0 and H=4 kOe. This indicates that the contribution from carriers of heat current except electrons to  $\kappa$  increases below  $T_c$  in the zero magnetic field because in this temperature region electrons are dominated by the elastic scattering as mentioned above. Considering that this enhancement of  $\kappa$ below  $T_c$  is suppressed by introducing vortices and the temperature dependence of  $\kappa$  under H=4 kOe is similar to that of the convensional superconductor whose heat current is carried mainly by electrons, it is natural to ascribe the origin of the enhancement of  $\kappa$  below  $T_c$  in the zero magnetic field to the increase of the phonon contribution. We simply assume that the total thermal conductivity is expressed as the sum of the phonon and electron contributions, i.e.,  $\kappa_{\mathrm{tot}} = \kappa_{\mathrm{ph}} + \kappa_{e}$  . The temperature dependence of the difference of the temperature dependence of  $\kappa$  normalized at  $T_c(H)$ between under H=0 and 4 kOe shown in the inset in Fig. 2 originates from  $\kappa_{\rm ph}$ .  $\kappa$  under H=4 kOe mainly originates from  $\kappa_e$ . In the zero magnetic field, the phonon mean free path becomes large below  $T_c$  due to the decrease of the number of normal electrons which act as scattering centers of phonons above  $T_c$ . This increase of the phonon mean free

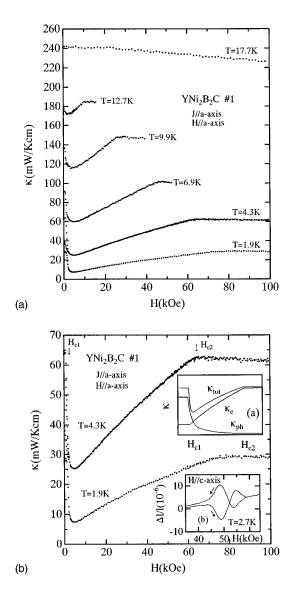


FIG. 3. Magnetic-field dependence of the thermal conductivity with heat current J parallel to the a axis of  $YNi_2B_2C$  sample 1 under the longitudinal magnetic field (a) in a temperature region up to 18 K and (b) in a low-temperature region below 4.3 K.  $H_{c1}$  and  $H_{c2}$  are the lower and upper critical field. Inset (1) shows a schematic picture of  $\kappa_{tot}$ ,  $\kappa_{ph}$ , and  $\kappa_e$ . See the text in detail. Inset (2) shows the longitudinal magnetostriction along the c axis in a magnetic field region around  $H_{c2}$ .

path causes the additional contribution to  $\kappa_e$  below  $T_c$  and this suggests the existence of a large electron-phonon coupling in this material. It is noted that such an enhancement of  $\kappa$  below  $T_c$  is not observed in  $\mathrm{Ba}_{1-x}\mathrm{K}_x\mathrm{BiO}_3$  with a relatively high  $T_c$  which is considered as the phonon-mediated strong-coupling superconductor. 12

Next, we discuss the magnetic-field dependence of  $\kappa$ . The phonon thermal conductivity  $\kappa_{\rm ph}$  enhanced below  $T_c$  in the zero magnetic field is largely suppressed by vortices introduced at  $H > H_{c1}$  because the phonon mean free path decreases as a result that phonons are scattered by vortices. In turn, the electron contribution becomes important with further increase of magnetic field. The schematic picture of the magnetic-field dependence of  $\kappa_{\rm ph}$  and  $\kappa_e$  together with  $\kappa_{\rm tot}$  below  $T_c$  is shown in inset (1) of Fig. 3(b). At low tempera-

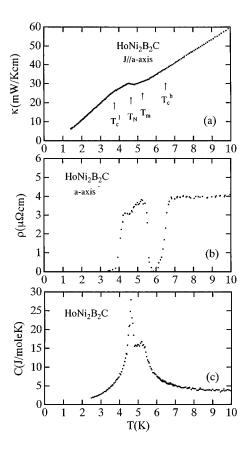


FIG. 4. Temperature dependence of (a) the thermal conductivity, (b) electrical resistivity, and (c) specific heat of  $\text{HoNi}_2\text{B}_2\text{C}$  at low temperatures.  $T_c^h$  and  $T_c^l$  are the higher and lower superconducting transition temperatures, respectively, and  $T_m$  and  $T_N$  are the incommensurate and commensurate antiferromagnetic transition temperatures, respectively.

tures, the negative curvature of the  $d\kappa/dH$  vs H curve is observed, which suggests the possible existence of lowenergy excitations in the superconducting state. The temperature dependence of  $\kappa$  under the magnetic field of 4 kOe shows  $\sim T^2$  behavior in a wide temperature region below  $T_c$ . At present, it is difficult to separate the electron contribution  $\kappa_e$  from  $\kappa_{\rm tot}$  in the superconducting state. Further studies are necessary to understand the magnetic field and temperature dependence of  $\kappa$  below  $T_c$ .

The magnetization curve shows the peak effect just below  $H_{c2}$  (Ref. 13) which are also observed in  $UPd_2Al_3$  and  $CeRu_2$  (Ref. 14) and has attracted much attention in relation to the possible existence of the Fulde-Ferrell-Larkin-Ovchinnikov state. Inset (2) of Fig. 3(b) shows the longitudinal magnetostriction under the magnetic field parallel to the c axis. This shows the pronounced anomaly in the same magnetic-field region as the peak effect in the magnetization curve is observed, as is observed in  $UPd_2Al_3$  and  $UPd_2Al_3$  and  $UPd_3Al_3$  and  $UPd_3Al_3$  and  $UPd_3Al_3$  is observed in  $UPd_3Al_3$  and  $UPd_3Al_3$  and

Figures 4(a), 4(b), and 4(c) show the temperature dependence of the thermal conductivity, electrical resistivity, and specific heat of HoNi<sub>2</sub>B<sub>2</sub>C, respectively. The magnetic properties of this compound were reported in Ref. 15. With the decrease of temperature, the electrical resistivity becomes

nearly zero at the higher superconducting transition temperature  $T_c^h \sim 6.6$  K and after recovering to the normal state value at ~5.3 K, it becomes zero at the lower superconducting transition temperature  $T_c^l \sim 3.6$  K. A small but clear decrease of  $\rho$  is observed at 4.6 K. The specific heat shows a small peak at  $T_m = 5.3$  K and a large and sharp peak at  $T_N = 4.6$  K which correspond to the incommensurate and commensurate antiferromagnetic ordering temperature (Ref. 15), respectively. The large short-range order effect is observed. These results are similar to the reported ones (Refs. 16 and 17) apart from slightly lower transition temperatures in the present sample. 18 At high temperatures, the anisotropic behavior of  $\kappa$  is observed as in YNi<sub>2</sub>B<sub>2</sub>C and the temperature dependence of  $L/L_0$  is also similar to those of YNi<sub>2</sub>B<sub>2</sub>C. However, low-temperature behavior of  $\kappa$  is different. At higher superconducting transition temperature  $T_c^h = 6.6$  K, we could not observe an anomaly in  $\kappa$ . A small but clear increase is observed in  $\kappa$  at  $T_N$ . This increase of  $\kappa$  originates from the decrease of  $\rho$  accompanying with the decrease of the magnetic scattering of electrons in the antiferromagnetic state. The clear decrease is observed in  $\kappa$  below  $T_c^l \sim 3.6$  K. At present, we do not know whether the heat current is dominated by phonons or electrons in the superconducting state in this compound. In order to clarify it, further studies are necessary at lower temperatures and under the magnetic field.

In conclusion, we have studied the thermal conductivity  $\kappa$  of high-quality YNi<sub>2</sub>B<sub>2</sub>C and HoNi<sub>2</sub>B<sub>2</sub>C single crystals. The results of YNi<sub>2</sub>B<sub>2</sub>C show the followings. The heat current is mainly carried by electrons at low temperatures in the normal state and in the superconducting state, is mainly carried by phonons which may be due to the increase of the phonon mean free path as a result of the decrease of the number of normal electrons below  $T_c$ . However, this phonon contribution is easily suppressed by introducing vortices at  $H > H_{c1}$  and the electron contribution becomes dominant above  $H_{c1}$ . The thermal conductivity of HoNi<sub>2</sub>B<sub>2</sub>C shows a clear increase just below the Néel temperature 4.6 K and a clear decrease below  $T_c^l \sim 3.6 \text{ K}.^{17,18}$ 

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