

# Effect of pressure on the superconducting properties and magnetism in $\text{HoNi}_2\text{B}_2\text{C}$

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## Abstract

The electrical resistance of a single crystal of  $\text{HoNi}_2\text{B}_2\text{C}$  has been measured as a function of temperature ( $T$ ) and magnetic field ( $H$ ) under high pressure ( $P$ ). As pressure increases,  $T_C$  decreases with the rate of  $\partial T_C/\partial P = -0.62$  K/GPa but  $T_N$  increases with  $\partial T_N/\partial P = 0.41$  K/GPa. This implies that the superconductivity above  $T_N$  competes with antiferromagnetism. The re-entrant superconducting temperature coexisting with the antiferromagnetic phase is increased by applying pressure. The present result indicates that  $T_C$  coincides with  $T_N$  near 3.2 GPa.

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## 1. Introduction

The family of the rare earth borocarbides having the formula  $\text{RNi}_2\text{B}_2\text{C}$  (R: rare earth elements) contains a few representatives of the magnetic superconductors [1]. For R = Tm, Er, Ho and Dy, the coexistence between superconductivity and antiferromagnetic ordering has been observed at low temperatures. The structure is tetragonal  $\text{ThCr}_2\text{Si}_2$  type, which consists of alternating layers of HoC planes and  $\text{Ni}_2\text{B}_2$  slabs. For  $\text{HoNi}_2\text{B}_2\text{C}$ , the two transition temperatures lie in the same temperature range 5–8 K. This compound has two different superconducting phases; the one co-exists with the antiferromagnetic order and the other nonmagnetic superconducting phase. It has been reported that  $T_C$  decreases and  $T_N$  increases by applying pressure up to 2 GPa [2,3]. This means that the pressure effect on the two superconducting phases are different, i.e., the superconductivity in antiferromagnetic phase becomes stable but the nonmagnetic one unstable

under pressure. This result suggests that  $T_C$  coincides with  $T_N$  at high pressure.

In the present work, we made an attempt to measure the temperature-dependent electrical resistance of a single crystal of  $\text{HoNi}_2\text{B}_2\text{C}$  up to 3 GPa, in order to examine the interplay of the superconductivity and antiferromagnetism at high pressure.

## 2. Experimental procedures

The samples were grown using the  $\text{Ni}_2\text{B}$  flux technique [1]. The electrical resistance was measured by using standard dc four-probe method. The current was in the  $ab$ -plane. A magnetic field was applied parallel to  $ab$ -plane using superconducting magnet. Hydrostatic pressure up to 3 GPa was generated using piston-cylinder method.

## 3. Experimental results

Fig. 1 shows the temperature-dependent electrical resistivity  $\rho(T)$  at various pressures up to 3 GPa. The resistance at

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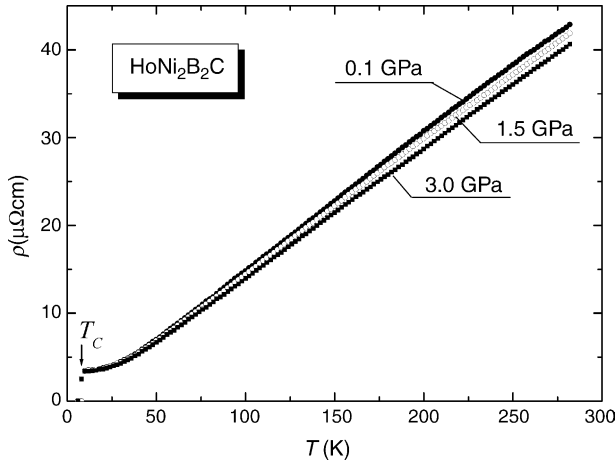


Fig. 1. Temperature dependence of the electrical resistivity of  $\text{HoNi}_2\text{B}_2\text{C}$  at high pressures up to 3 GPa.

room temperature decreases with increasing pressure with the rate  $(1/R)\partial R/\partial P = -1.7 \times 10^{-2} \text{ GPa}^{-1}$ . The  $\rho(T)$  curves show a smooth decrease with decreasing temperature without any discontinuities, followed by a sudden decrease at  $T_C$ , due to a superconducting transition.

Fig. 2 shows a detail of the  $\rho(T)$  curves at low-temperature below 10 K at various pressures. At 0.1 GPa, the superconducting transition begins around 9.2 K and is complete at 8.8 K. Above 2.5 GPa a re-entrant peak is observed in  $\rho(T)$  around 6 K, while it is not seen below 2.5 GPa. The maximum of  $\rho(T)$  is enhanced by applying pressure more but is suppressed near 3 GPa. This behavior is considered to be due to the competing pressure effect between the nonmagnetic superconducting phase and magnetic ordered phase.

We defined five characteristic temperatures, the onset of transition as (1)  $T_C$  (onset), the offset of transition as (2)  $T_C$  (offset), a maximum of the re-entrant peak as (3)  $T_N$  and the temperatures showing  $R = 0$  in the re-entrant phase as (4)

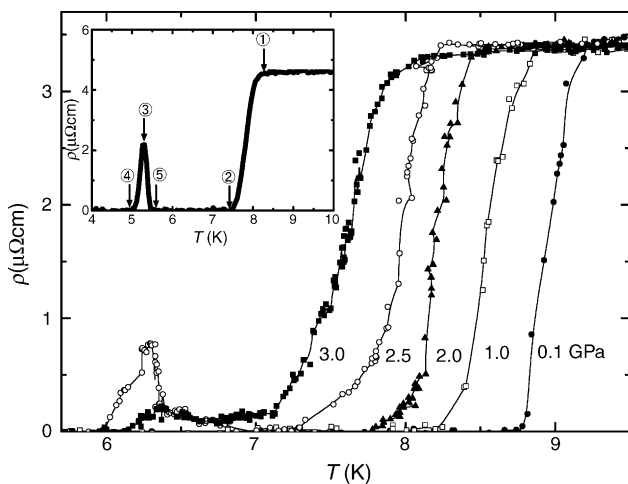


Fig. 2. Temperature-dependent electrical resistivity between 5.5 and 9.5 K at high pressures. The arrows in the inset: (1)  $T_C$  (onset), (2)  $T_C$  (offset), (3)  $T_N$ , (4)  $T_{C1}^{\text{AF}}$  and (5)  $T_{C2}^{\text{AF}}$ .

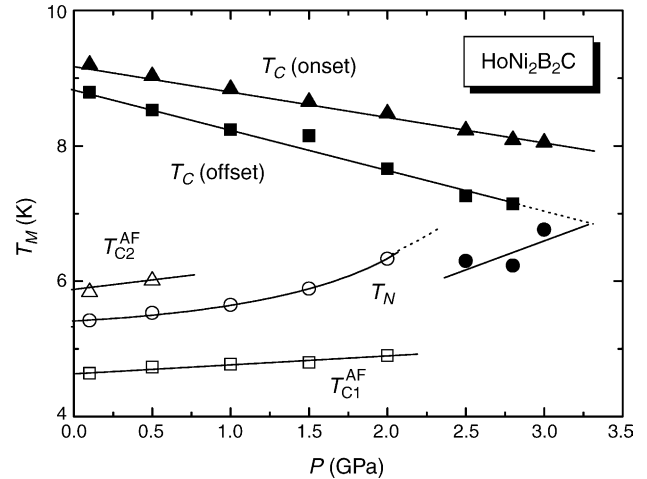


Fig. 3. Pressure dependence of  $T_C$  (onset),  $T_C$  (offset),  $T_{C1}^{\text{AF}}$ ,  $T_{C2}^{\text{AF}}$  and  $T_N$ . Open circles show  $T_N$  at a magnetic field  $H = 1.5 \text{ kOe}$  and filled circles show  $T_N$  at zero field. Filled triangles and squares show  $T_C$  (onset) and  $T_C$  (offset) at zero field. Open squares and triangles show  $T_{C1}^{\text{AF}}$  and  $T_{C2}^{\text{AF}}$  at a magnetic field  $H = 1.5 \text{ kOe}$ , respectively. The solid lines are guide of eye.

$T_{C1}^{\text{AF}}$  and (5)  $T_{C2}^{\text{AF}}$ , the former is below  $T_N$  and the latter above  $T_N$ , respectively. These are shown in the inset of Fig. 2. As pressure increases,  $T_C$  (onset) and  $T_C$  (offset) decrease but  $T_N$  increases, and the superconducting transition no longer completes above  $T_N$  at 3 GPa because a finite resistivity remains between 6.7 and 7.1 K. This fact means that there is no paramagnetic superconducting state above 3 GPa. It should be noted that the shape of  $\rho(T)$  curves is changed by increasing pressure; at low pressures (below 2 GPa) the resistivity becomes zero sharply but by further pressurizing, a tail just above  $T_C$  (offset) is developed and the width of the transition becomes large.

Fig. 3 displays the pressure dependence of  $T_C$  (onset),  $T_C$  (offset),  $T_{C1}^{\text{AF}}$ ,  $T_{C2}^{\text{AF}}$  and  $T_N$ . Below 2.5 GPa, the re-entrant peak in the  $\rho(T)$  curve does not appear at  $H = 0$ , but is seen at small applied magnetic field,  $H = 1.5 \text{ kOe}$  [4].  $T_N$ ,  $T_{C1}^{\text{AF}}$  and  $T_{C2}^{\text{AF}}$  are obtained at  $H = 1.5 \text{ kOe}$ . Above 2.5 GPa, it was difficult to obtain  $T_N$  from the re-entrant peak at 1.5 kOe because the peak in  $\rho(T)$  due to antiferromagnetic ordering merged into the superconducting transition. So we use the data of  $T_N$  at 1.5 kOe below 2.5 GPa and  $T_N$  at  $H = 0$  above 2.5 GPa. The magnetic field dependence on  $T_N$  was negligibly small. At 3 GPa, the transition did not complete but the temperature extrapolated to  $R = 0$  ( $\sim 7 \text{ K}$ ) is slightly above  $T_N$ .  $T_C$  (offset) and  $T_N$  are close each other, which gives rise to a strong interaction between superconductivity and antiferromagnetism. Both  $T_C$  (onset) and  $T_C$  (offset) decrease with increasing pressure with the rates,  $\partial T_C(\text{onset})/\partial P = -0.40 \text{ K/GPa}$  and  $\partial T_C(\text{offset})/\partial P = -0.62 \text{ K/GPa}$ , respectively.  $T_N$  increases with pressure,  $\partial T_N/\partial P = +0.41 \text{ K/GPa}$ . Compared with the previous data [2,4],  $\partial T_C(\text{offset})/\partial P = -0.46$  to  $-0.50 \text{ K/GPa}$  and  $\partial T_N/\partial P = 0.70\text{--}0.84 \text{ K/GPa}$ , the present results show that the pressure derivative of  $T_C$  (offset) is nearly the same but that of  $T_N$  is different. The origins of

the difference are partly due to an experimental error but not clear now. There are also sample dependence of pressure dependence of  $T_N$  and  $T_C$ .

## 4. Discussion

### 4.1. Temperature dependence of $\rho$ above $T_C$ (onset)

To examine the temperature dependence of electrical resistivity above  $T_C$  (onset), we assume the following power law [5]:

$$\rho(T) = \rho_0 + AT^n, \quad (1)$$

where  $\rho_0$  and  $A$  are the residual resistivity and the constant depending on pressure. Fig. 4 shows the plot of  $(\rho(T) - \rho_0)$  as a function of  $T$  at 0.1 and 2.5 GPa in the logarithmic scale, in which the gradients correspond to the value of  $n$ .  $\rho_0$  is estimated by extrapolating resistivity above  $T_C$  (onset) to  $T = 0$ . It is found that there is no difference between 0.1 and 2.5 GPa within experimental error and we obtain  $n = 2.0 \pm 0.2$ . This result indicates the electronic state above  $T_C$  (offset) is described by normal Fermi liquid, which is stable under high pressure at least up to 3 GPa. The values of  $\rho_0$  are also nearly constant below 3 GPa. The value of  $A$  is about  $8.0 \times 10^{-4} \mu\Omega \text{ cm K}^{-2}$ , which is almost constant against pressure. This result means the effect of pressure on spin fluctuation is small because  $A$  is related to the magnitude of spin fluctuation. The magnitude of  $A$  is smaller than the value  $A = 3.0 \times 10^{-3} \mu\Omega \text{ cm K}^{-2}$ , which is expected from Kadowaki–Woods plot and by using the value  $\gamma = 17.5 \text{ mJ/mol K}^2$  [6,7].

The coefficient  $A$  is proportional to the square of the density of state of conduction electrons at Fermi level  $N(0)$  [8].

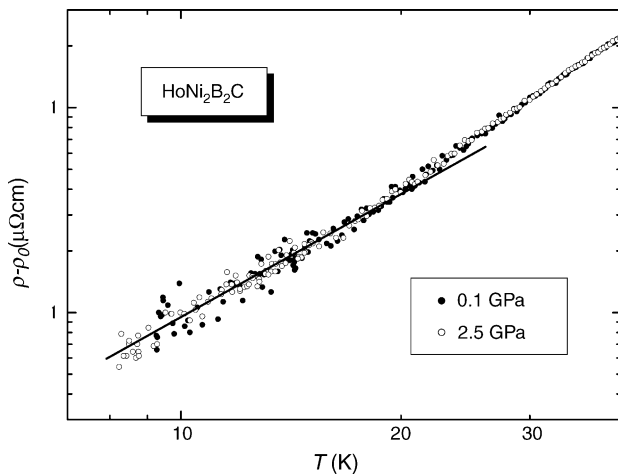


Fig. 4.  $(\rho(T) - \rho_0)$  as a function of  $T$  at 0.1 and 2.5 GPa at zero field in logarithmic scale. Filled and open circles show the data at 0.1 and 2.5 GPa, respectively.

$T_C$  is represented by following equation:

$$T_C = 1.14\Theta_D \exp\left(-\frac{1}{N(0)V_{\text{ph}}}\right), \quad (2)$$

where  $\Theta_D$  and  $V_{\text{ph}}$  are Debye temperature and electron-phonon interaction energy, respectively. The volume dependence of  $T_C$  is described as

$$\begin{aligned} \frac{\partial \ln T_C}{\partial \ln V} &= \frac{\partial \ln \Theta_D}{\partial \ln V} + \ln\left(\frac{1.14\Theta_D}{T_C}\right) \frac{\partial \ln N(0)}{\partial \ln V} \\ &+ \ln\left(\frac{1.14\Theta_D}{T_C}\right) \frac{\partial \ln V_{\text{ph}}}{\partial \ln V}. \end{aligned} \quad (3)$$

Since  $A$  and  $\Theta_D$  are almost independent of pressure as mentioned in the foregoing paragraph, the first and second term may be 0. We obtained  $\Gamma[T_C(\text{offset})] = -\partial \ln T_C(\text{offset})/\partial \ln V = -13.6$ , which is relatively large compared with normal superconductors [9]. Taking into account these results, the pressure dependence of  $T_C$  is originated mainly from the pressure effect on electron-phonon interaction. The value of  $(1/V_{\text{ph}})\partial V_{\text{ph}}/\partial P$  is estimated to be  $-1.9 \times 10^{-2} \text{ GPa}^{-1}$  by using Eq. (3), which is the same order of magnitude as that of  $\text{TmNi}_2\text{B}_2\text{C}$  [10].

### 4.2. Coexistence of antiferromagnetism and superconductivity

The re-entrant peak appears above 2.5 GPa at  $H = 0$  but it is not seen below it. This is caused by the antiferromagnetic correlation of 4f-electrons enhanced by applying pressure to give magnetic interaction to overcome the superconductivity. Compressibility of this compound has large anisotropy; linear compressibility of  $c$ -axis is two times larger than that of  $a$ -axis [4]. This fact implies that the distance between layers shrinks more than that in the layers. Consequently, the application of pressure strengthens antiferromagnetic correlation but weakens the superconductivity in nonmagnetic phase.

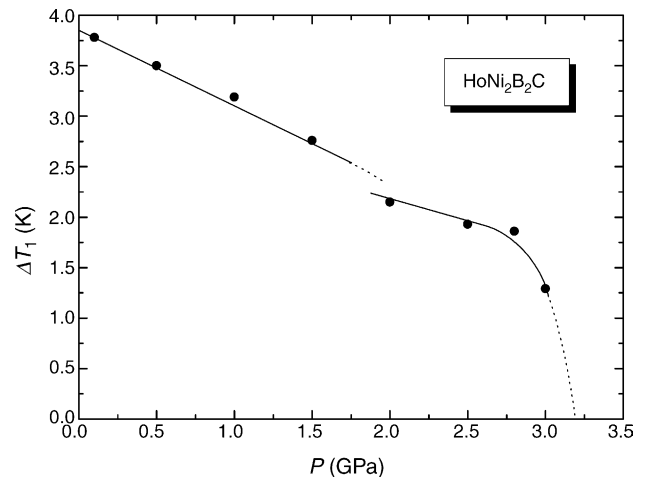


Fig. 5.  $\Delta T_1 = T_C(\text{onset}) - T_N$  as a function of pressure.

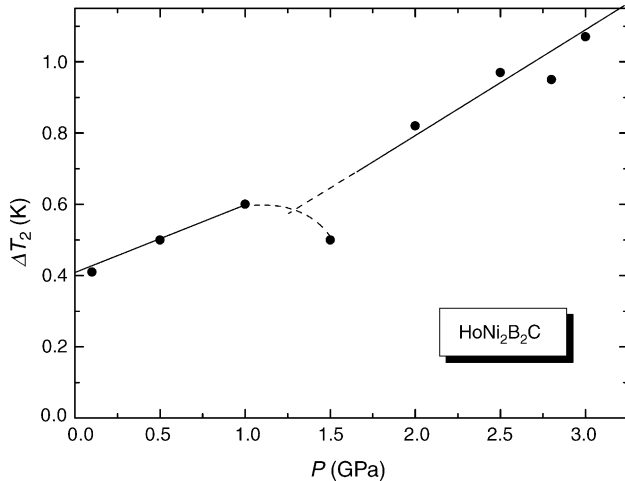


Fig. 6. Pressure dependence of the width of superconducting transition,  $\Delta T_2 = T_C(\text{onset}) - T_C(\text{offset})$ .

Measurements of  $H_{C2}$  indicate that there is a drastic dip in  $H_{C2}$  data around  $T_N$  [1]. A previous study reported that the dip of  $H_{C2}$  smears to zero by applying pressure [11]. The sudden appearance of the re-entrant peak at 2.5 GPa is due to the fact that pressure dependence of  $T_N$  is larger than that of  $T_{C1}^{\text{AF}}$  as shown in Fig. 3.

To investigate the effect of pressure on  $T_C$  and  $T_N$ , we define,  $\Delta T_1 = T_C(\text{onset}) - T_N$ ,  $\Delta T_1$  is plotted in Fig. 5 as a function of pressure. The pressure dependence of  $\Delta T_1$  is found to change in near 2 GPa. This is caused by a rapid increase of  $T_N$  with increasing pressure above 2 GPa. Considering that the re-entrant peak appears above 2 GPa, the antiferromagnetic interaction among  $\text{Ho}^{3+}$  ions may be especially enhanced by pressure around 2 GPa, i.e., the interaction between  $T_C$  (onset) and  $T_N$  may become strong as both temperatures are close.  $\Delta T_1$  is extrapolated to 0 around 3.2 GPa.

The width of superconducting transition,  $\Delta T_2$ , defined as  $\Delta T_2 = T_C(\text{onset}) - T_C(\text{offset})$  is plotted in Fig. 6.  $\Delta T_2$  in-

creases linearly with pressure but the point at 1.5 GPa deviates downward. Then it increases again by further pressurizing. It is caused by a tail on the superconducting transition just above  $T_C$  (offset). Pressure inhomogeneity sometimes makes the width of the transition broad. But the tail in  $\rho(T)$  at 3 GPa is shorter than that at 2.5 GPa. Considering this fact, it should be ruled out as an origin of long tail. This long tail is reminiscent of behavior of vortex at magnetic field in another borocarbide compound such as  $\text{YNi}_2\text{B}_2\text{C}$  [12]; a magnetic field changes the vortex solid due to field-enhanced pinning. The anomalous behavior above 2 GPa suggests that pressure affects a motion of vortex in a similar fashion as the effect of a magnetic field mentioned above. But the details of mechanism of the tail above 2.5 GPa are not known at present.

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