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The competition between superconductivity and antiferromagnetism in $Y_{1-x}Tb_xNi_2B_2C$ single crystals

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Abstract

Magnetic and superconducting properties in a series of intermetallic compounds $Y_{1-x}Tb_xNi_2B_2C$ were investigated by measuring the temperature dependent magnetization, $M(T)$, and resitivity, $\rho(T)$. As Tb concentration, x, is increased, the superconducting transition temperature, T_C, decreases and eventually disappears in the vicinity of x = 0.5 while Néel temperature, T_N, appears abruptly near x = 0.4 and increases linearly. Of particular interest is the collision of superconductivity and antiferromagnetism around x = 0.4. The linear decrease of T_C for dilute Tb concentration seems to follow the Abrikosov-Gor'kov behavior, while the decay of T_C below T_N is expected to originate from the effective magnetic field on the conduction electrons. The Ginzburg-Landau theory was phenomenologically constructed to explain this competition of superconducting order parameter and antiferromagnetic order parameter with the multi-band model.

1. Introduction

The quaternary borocarbides RNi_2B_2C (R = Y, rare-earth elements) have been continuously studied because of their comparatively high superconducting transition temperatures, up to T_C = 23 K, and the coexistence of superconductivity and magnetism. Even though they have a layered tetragonal structure with alternating RC sheets and Ni_2B layers[1], similar to high T_C cuprate superconductors, electronic band calculations for $LuNi_2B_2C$ demonstrated that they are electronically three dimensional. It was also known that Ni 3d electrons predominantly contribute to the conduction band, along with other elements B and C.[2]

For pure quaternary compounds, T_C scales well with the de Gennes (dG) factor according to the Abrikosov-Gor'kov theory.[3] However, this is no more valid for $Ho_{1-x}Dy_xNi_2B_2C$ and $Lu_{1-x}Dy_xNi_2B_2C$.[4] In $Ho_{1-x}Dy_xNi_2B_2C$, which exhibits $T_C > T_N$ at x = 0 and $T_C < T_N$ at x = 1, T_C drops with the increase of the dG factor for $T_C > T_N$, while T_C is independent of the dG factor for $T_C < T_N$. Since Ho and Dy spins with the similar magnitude have almost the same commensurate antiferromagnetic structure, it is claimed that the effective magnetic field at the Ni site is completely canceled and the superconductivity can survive below T_N. That might be also the reason why T_C does not vary in the antiferromagnetic (AF) state of $Ho_{1-x}Dy_xNi_2B_2C$. In contrast, T_N is proportional to the dG factor, as expected from the Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism. On the other hand, in $Lu_{1-x}Dy_xNi_2B_2C$, T_C is demolished for x < 0.4 and T_C is restored below T_N for x > 0.5, as in Fig.5. In Dy-rich case, Lu^{+3} may hinder the net magnetic field produced by the nearest Dy spins from being completely canceled at the Ni site and cause magnetic pair–breaking effect despite nonmagnetic ion.[5]

In this paper, we report a systematic study of the superconductivity and magnetism in $Y_{1-x}Tb_xNi_2B_2C$ single crystals, composed of nonmagnetic Y (T_C = 16.5 K) and nonsuperconducting Tb (T_N = 15 K). The phenomenological model based on the Ginzburg-Landau theory is adopted to explain the coexistence of superconductivity and antiferromagnetism.

2. Experiment

Sizable single crystals of $Y_{1-x}Tb_xNi_2B_2C$ were grown by flux method using Ni_2B as a solvent.[6]

DC magnetization measurements were performed using a Quantum Design SQUID magnetometer. The zero-field electrical resistivity was measured by standard four-probe method.

3. Results

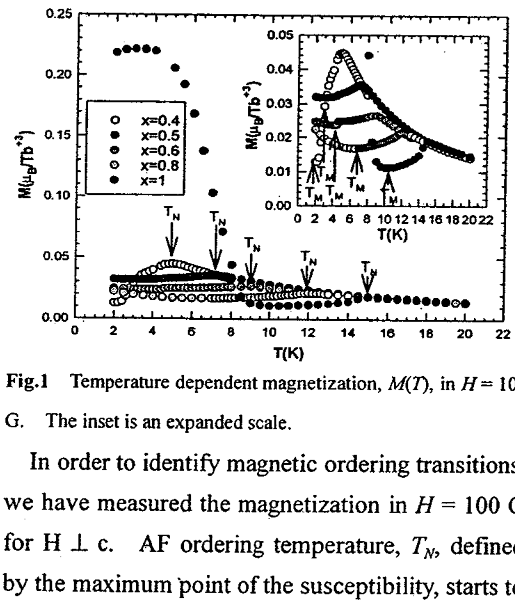

Fig.1 Temperature dependent magnetization, $M(T)$, in H = 100 G. The inset is an expanded scale.

In order to identify magnetic ordering transitions, we have measured the magnetization in H = 100 G for H ⊥ c. AF ordering temperature, T_N, defined by the maximum point of the susceptibility, starts to be revealed from x = 0.4 and is obviously increased with doping Tb, as in Fig.1. Moreover, T_N has a roughly linear dependence on the dG factor. In particular, for x = 0.4, T_N is so close to T_C that it is difficult to be discriminated. Whereas, in Fig.1, anomalous behavior in $M(T)$ is observed at low temperatures, which develops with the increase of Tb content. As can be seen in the inset of Fig.1, onset of the increase in $M(T)$ at lower temperature is regarded as weak ferromagnetic ordering temperature, T_M. In the case of $TbNi_2B_2C$, magnetization is drastically raised below $T \approx 10$ K and saturated below $T \approx 4$ K. The saturated moment at T = 2 K is approximately 0.22 μ_B.[6] As x is increased, the increase in $M(T)$ becomes more apparent and T_M is enhanced.

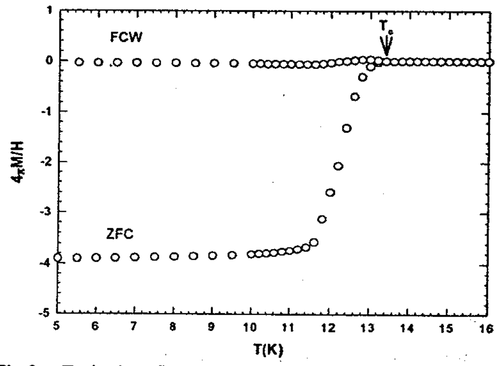

Fig.2 T_C in low-field temperature dependent susceptibility, $4\pi M/H(T)$, for both ZFC and FCW process at x = 0.1.

To investigate the superconductivity of these materials, low field magnetization was measured for H ll c. In Fig.2, for example of x = 0.1, zero field cooled (ZFC) magnetization is remarkably lower than field cooled (FC) case and shows distinct diamagnetism at temperatures below 13.4 K. T_C was determined by the onset of the diamagnetic drop of ZFC magnetization. In ZFC case, a volume fraction for shielding is more than 100 % due to the demagnetization effect.

In Fig.3, the relative in-plane electrical resistivity as a function of temperature in the zero field shows that a sharp superconducting transition above T = 2 K exists up to x = 0.4 and completely vanishes above x = 0.5. The rapid decline of T_C with increasing Tb content is similar to the results from the low-field magnetization measurement.

These temperatures, T_C, T_N and T_M as a function of the dG factor are all exhibited in Fig.4. As Tb concentration, x, is increased, T_C is reduced and disappears near x = 0.5, while T_N is linearly increased above x = 0.4 and T_M is enhanced above x = 0.4 at lower temperature.

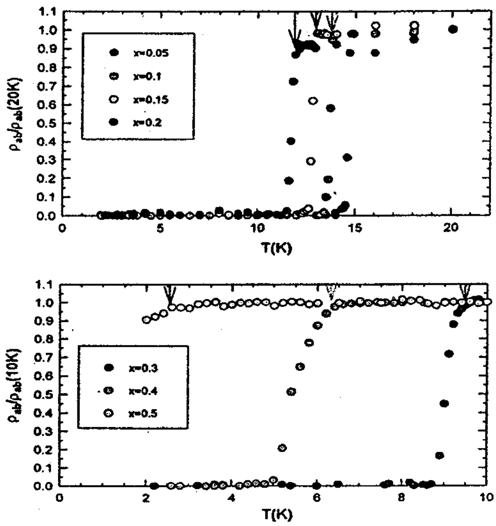

Fig. 3 Resistivity in the ab plane, ρ_{ab}, vs. T for x = 0.05 – 0.5. The arrow indicates the onset of the superconducting transition for each x, defined as T_C.

4. Discussion

The behavior of T_C and T_N in the phase diagram of Fig.4 can be described by the competition of the superconducting order and the AF order, as mentioned above. Now we construct the homogeneous Ginzburg-Landau free energy, which contains both the order parameters as follows.

$$F = a_S(T - T_{C0})|\phi|^2 + \frac{b_S}{2}|\phi|^4 + a_M(T - T_{N0})M^2 + \frac{b_M}{2}M^4 + \gamma M^2|\phi|^2 \quad (1)$$

where ϕ and M indicate the superconducting and AF order parameters, respectively. T_{C0} represents the superconducting transition temperature of pure YNi_2B_2C, and T_{N0} is Néel temperature and proportional to the dG factor. The coupling constant of the superconducting order and AF order, γ, is assumed to depend on the dG factor, as is the case of the Abrikosov-Gor'kov pair breaking theory. Owing to the interaction of the two order parameters, the real T_C and T_N will be suppressed from its original values, T_{C0} and T_{N0}.

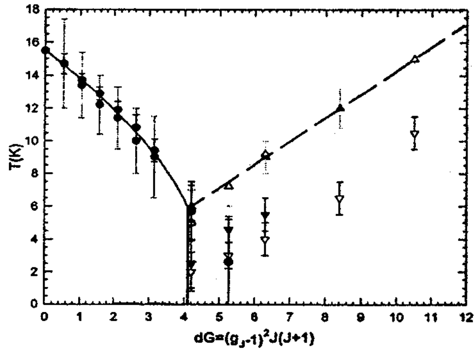

Fig.4 T_C vs. dG factor from $R(T)$(solid circle), from $M(T)$ in H = 10 G(gray circle), T_N from $M(T)$ in H = 10 G(solid up-triangle), in H = 100 G(open up-triangle) and T_M from $M(T)$ in H = 10 G(solid down-triangle), in H = 100 G(open down-triangle). The solid line denotes T_C from GL theory and the dashed line, T_N from GL theory.

To explain the T_C suppression more rigorously, we need to consider the fluctuation effects, but since this effect in superconductivity is small, we considered only the magnetic fluctuation effect. We separate the AF order parameter into its mean value $\overline{M}$ and fluctuation δM. The free energy change due to the magnetic fluctuation with the mean field approximation of δM^2 can be written as

$$\delta F \cong a_M(T - T_{C0})\delta M^2 + b_M\langle\delta M^2\rangle\delta M^2 + 3b_M\overline{M}^2\delta M^2 + \gamma|\varphi|^2\delta M^2 \quad (2)$$

Using the Gaussian fluctuation, we get the self-consistent field equation as follows.

$$\langle\delta M^2\rangle = \frac{k_B T}{2(a_M(T - T_{N0}) + 3b_M\overline{M}^2 + \gamma|\phi|^2 + b_B\langle\delta M^2\rangle)} \quad (3)$$

By minimizing the free energy Eq.(1) with respect to ϕ and $\overline{M}$, two more equations are obtained.

$$|\phi|^2 = -\frac{a_S(T - T_{C0}) + \gamma\langle\delta M^2\rangle + \gamma\overline{M}^2}{b_S},$$

$$\overline{M}^2 = -\frac{a_M(T-T_{N0})+\gamma|\phi|^2}{b_M} \qquad (4)$$

With the temperature dependence of $\langle \delta M^2 \rangle, \overline{M}^2$ and $|\phi|^2$ from Eq.(3), Eq.(4), the reduced T_C and T_N can be acquired by solving

$$a_S(T-T_{C0})+\gamma\langle \delta M^2 \rangle+\gamma\overline{M}^2 = 0 \qquad (5)$$

$$a_M(T-T_{N0})+\gamma|\phi|^2 = 0 \qquad (6)$$

with respect to T, respectively. Tb concentration, x, will modify T_{N0} and γ through the change of the effective dG factor. Now we get the calculated results of T_C and T_N vs. x in Fig.4.

The linear dependence of T_N in the experiment verifies our assumption that the superconducting fluctuation is small. The AF order is affected only near x = 0.4 where the T_C and T_N meet and the superconducting fluctuation exists, while the T_C curve between x = 0.35 and x = 0.4 deviates from the linear behavior due to the magnetic fluctuation. Our calculated results show no superconductivity when the antiferromagnetism exists, and vice versa. The experiments, however, show both T_C and T_N at x = 0.4 and 0.5. According to Fig.1, the AF transitions of both the samples in *M/H* (T) curve are rather broad than that of pure $TbNi_2B_2C$. This tells us that the inhomogeneity of the AF order which we didn't consider in the model could be responsible for their coexistence. Or the magnetic structure modified from that of the pure Tb sample due to the competition with the superconductivity, may go into the state where the superconductivity can survive. For x < 0.4, the magnetic rare earth atoms are so dilute that its magnetic structure can be easily disturbed. In contrast, for x > 0.6, superconductivity cannot exist below T_N because of strong pair breaking field on conduction electrons.

5. Summary

To better understand the competition between superconductivity and magnetism in borocarbides, we performed the magnetization and zero-field resistivity measurements on $Y_{1-x}Tb_xNi_2B_2C$ single crystals. For dilute Tb concentration, T_C looks as if it follows the Abrikosov-Gor'kov theory, but diminishes rapidly with the emergence of T_N. T_N has a linear dependence on the dG factor. No superconductivity in $Y_{1-x}Tb_xNi_2B_2C$ above x = 0.5, as well as in $TbNi_2B_2C$, is preferably expected to originate from the strong pair-breaking field due to noncollinear AF structure of Tb^{+3} ions below T_N. It was successfully explained by using the Ginzburg-Landau theory.

6. Acknowledgement

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7. Reference

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