Effect of Tb substitution for Dy on superconductivity and magnetism in $Dy_{1-x}Tb_xNi_2B_2C$

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The superconductivity and the magnetism of $Dy_{1-x}Tb_xNi_2B_2C$ ($0.0 \leqslant x \leqslant 1.0$) single crystals were studied. As Tb^{+3} was substituted for Dy^{+3}, the superconducting transition temperature T_c decreased linearly at a rate of $dT_c/dx = -12$ K, and for $x \geqslant 0.4$, no superconductivity was observed above 2 K. In contrast, the antiferromagnetic ordering temperature T_N was almost constant for $x \leqslant 0.6$. These behaviors are in sharp contrast to the case of $Dy_{1-x}Ho_xNi_2B_2C$, where the T_c is constant for $x < 0.7$ but the T_N decreases linearly with x. The difference between $Dy_{1-x}Tb_xNi_2B_2C$ and $Dy_{1-x}Ho_xNi_2B_2C$ can be understood if the magnetic structures of both compounds are properly considered.

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I. INTRODUCTION

A family of superconductors, RNi_2B_2C (R=rare-earth element, Y, and Lu),[1–3] shows many interesting features, such as the coexistence of superconductivity and magnetism,[2,3] the formation of a square vortex lattice,[4,5] a large fluctuation effect,[6–9] an upward curvature of the upper critical field,[10,11] and anomalous optical properties.[12,13] While the gap symmetry of this family of compounds was thought to be that of a fully gapped s wave, this issue was revisited recently, based on studies on the field-angle-dependent specific heat[14] and thermal conductivity,[15] and an anisotropic or nodal superconducting gap was proposed. This anisotropic or nodal superconducting gap was also found to coexist with nonlocal effects in $LuNi_2B_2C$, where a complex field-angle dependence of the specific heat appeared when the sample became disordered.[16] Among the issues surrounding this family of superconductors,[17] the interplay between magnetic ordering and superconductivity is quite interesting not only because the ranges of the superconducting and the antiferromagnetic (AF) critical temperatures T_c and T_N are very wide but also because the configuration of the magnetic structure varies differently depending on R.[18]

RNi_2B_2C has a layered structure with a tetragonal symmetry, which consists of $(Ni\text{-}B)_2$ and R-C layers along the c direction. Despite this layered structure, band structure calculations have showed that the electronic properties of these compounds are three dimensional.[19,20] In addition, this family of compounds has a mulitiband character; the electrons in the Ni $3d$ band are major charge carriers and are the main contribution to the superconductivity, even though the contributions from other bands are not negligible.

The low-temperature magnetic structures of RNi_2B_2C (Refs. 2 and 3) have been studied in detail by using neutron scattering.[21] For example, as Fig. 1 shows, in the magnetically ordered state, the magnetic spins for R=Dy (Ho) align in the [110] direction ferromagnetically in the ab plane but antiferromagnetically along the c direction. For R=Tb, however, the magnetic spins point along the a axis and form an a-axis-modulated ordered structure. This ordering of the magnetic spins of the R^{+3} ions is mediated by the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, which is an exchange interaction between the localized magnetic spins and the conduction electrons. The Néel temperatures T_N's of the pure quaternary compounds for R from R=Tm to Gd are nearly proportional to the de Gennes factor $dG = (g_J - 1)^2 J(J+1)$, where g_J is the Lande g factor, and J is the total angular momentum, which is determined by Hund's rules for the ground state of R^{+3}. The values of dG are 4.5, 7.1, and 10.5 for Ho^{+3}, Dy^{+3}, and Tb^{+3}, respectively and T_N increases in this order.

Since the magnetic moments perturb the singlet (spin up and spin down) Cooper pairs formed between the conduction electrons, the magnetic ion R^{3+} suppresses the superconductivity. The pair-breaking effect due to exchange scattering by *uncorrelated* local magnetic spins was theoretically considered by Abrikosov and Gor'kov (AG).[22] According to the AG theory, the T_c is rapidly suppressed with increasing dG;

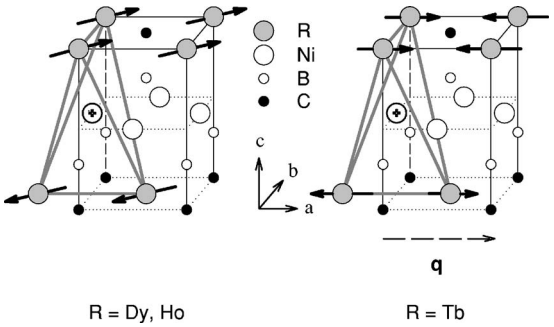

FIG. 1. Crystalline structure and magnetically ordered state of RNi_2B_2C at low temperatures. The arrow on a R ion represents its magnetic spin. The magnetic ordering wave vector for the Tb case is indicated by **q**.

i.e., $dT_c/dx \propto -J_{sf}^2 dG$, where x is the concentration of magnetic moments and J_{sf} is an exchange coupling constant between the local moments and the conduction electrons. In pure $R Ni_2B_2C$, where the magnetic elements R are located on regular lattice sites, the T_c decreases monotonically with increasing dG from R=Y, Lu to R=Dy.[18] It should be noted that in the case of R=Dy, superconductivity appears inside the magnetic order and in this case, AG theory is no longer valid. Also, for the alloy $(R_{1-x}R'_x)Ni_2B_2C$, the T_c is suppressed as the effective de Gennes factor, $dG_{eff}=(1-x)\times dG[R]+x\times dG[R']$, increases as long as the T_c is higher than the T_N,[23] as is predicted by the AG theory. However, T_c/T_{c0} versus the dG factor for these compounds did not show a universal behavior. This breakdown of dG scaling originated from different crystalline electric field (CEF) effects of R^{3+} and different values of conduction electron-local moment coupling constant.[23]

de Gennes scaling of the T_c was also found to break down in the $Dy_{1-x}Ho_xNi_2B_2C$ system when T_N was higher than T_c.[23,24] This anomalous behavior was not confined only to the Dy—Ho system. When nonmagnetic ions, such as Lu^{+3} (Ref. 23) and Y^{+3} (Ref. 25) are substituted for Dy^{+3}, the T_c is suppressed rapidly. On the other hand, as Ho^{+3} replaces Dy^{+3}, the T_N of the Dy—Ho system decreases linearly with decreasing dG_{eff}, as expected. Y substitution for Tb has also been studied in detail.[26,27]

To understand the above-mentioned phenomena more clearly, we prepared $Dy_{1-x}Tb_xNi_2B_2C$ (Dy—Tb) single crystals, in which T_c was lower than T_N. In that system, we found T_c and T_N behaviors that were strikingly different from those in the Dy—Ho system. In the Dy—Tb system, the T_c is depressed significantly by a small substitution of the magnetic ion Tb^{+3} while the T_N does not change very much for $x \leqslant 0.6$. This is an unexpected result in view of the T_c and the T_N behaviors in the Dy—Ho system. We found that the different magnetic structures for Tb and Dy (or Ho) were very crucial to understanding the strange pair-breaking and magnetic ordering.

II. EXPERIMENT

Crystals of $Dy_{1-x}Tb_xNi_2B_2C$ were prepared for ten different nominal compositions from x=0.0 to 1.0 by using a high-temperature metal flux of Ni_2B, as has been described in detail elsewhere.[28] The crystal structure of each composition was confirmed to be tetragonal, like that of the two mother compounds, by using powder x-ray diffraction. The temperature- and field-dependent magnetizations, as well as the resistances, were measured using a commercial superconducting quantum interference magnetometer. The zero-field-cooled (ZFC) data in the superconducting state were obtained on warming after a sample had been quenched.

III. RESULTS AND DISCUSSION

The temperature-dependent static susceptibilities χ of the $Dy_{1-x}Tb_xNi_2B_2C$ compounds with x=0.05, 0.1, and 0.25 under the zero-field-cooled (ZFC) condition for $\mathbf{H}\|\mathbf{c}$ are plotted in Fig. 2(a). To probe the superconducting transitions with the magnetic disturbances caused by the Dy^{+3} and the Tb^{+3} ions minimized, we applied a low magnetic field of 2 Oe along the c direction, which is the magnetic hard axis of these compounds. The $4\pi\chi$ for x=0.05 shows a typical superconducting diamagnetism, but the transition width ΔT_c of 2 K from the onset of the diamagnetic signal to 90% of the value at 2 K is somewhat broad. The value of $4\pi\chi \approx -4.6$ at T=2 K is larger than the 100% diamagnetic value due to the demagnetization effect. For x=0.25, the data, even at T=2 K, show that the samples are on the way to becoming superconductors with volume fractions of less than 100%, which indicates indirectly that the competition between superconductivity and magnetism exists. The T_c for each case was determined from the onset of diamagnetism and is designated by an arrow in Fig. 2(a). Substituting Tb for Dy in $DyNi_2B_2C$ surely weakens the superconductivity and reduces the T_c. For the samples with $x \geqslant 0.4$, the superconductivity was totally destroyed, which was observed in the magnetization measurements and in the electric resistance measurements shown in Fig. 2(b). In the resistance measurements, the sudden change in the temperature dependence above T_c was found to originate from the AF ordering. The values of T_c determined from the resistance measurements are nearly the same as those determined from $\chi(T)$.

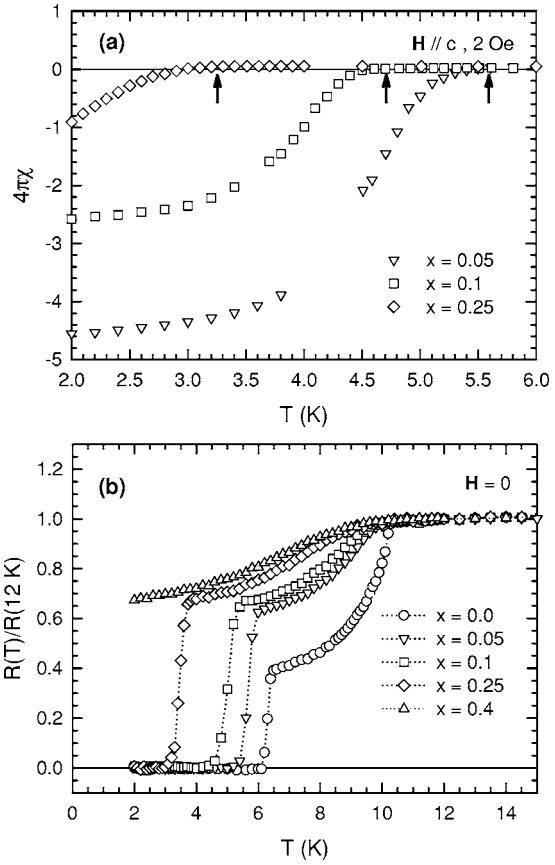

FIG. 2. (a) Static susceptibility of $Dy_{1-x}Tb_xNi_2B_2C$ with x =0.05, 0.1, and 0.25 for $H\|c$ versus temperature in a zero-field-cooled warming condition. The arrows indicate the superconducting transition temperatures. (b) Normalized resistances of $Dy_{1-x}Tb_xNi_2B_2C$ with x=0.05, 0.1, and 0.25 versus temperature. Because of the AF ordering, the resistances suddenly decrease at $T=T_N$.

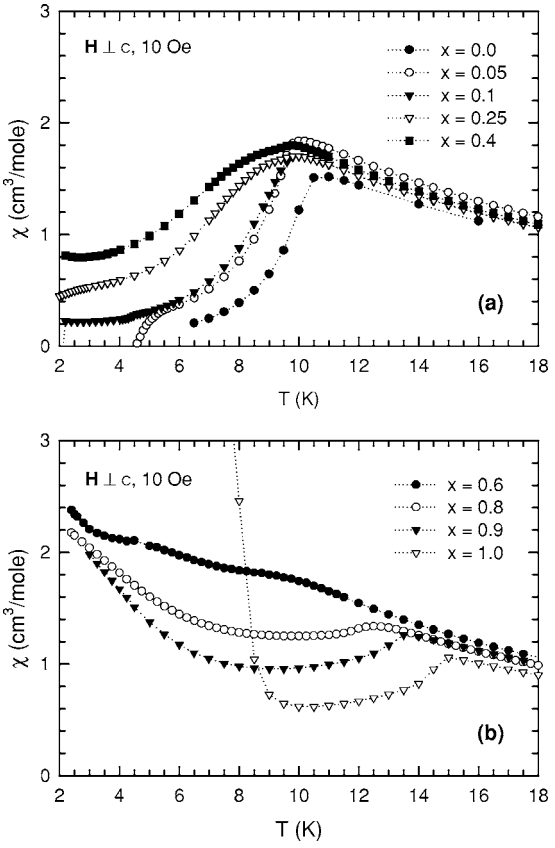

FIG. 3. Static susceptibility $\chi(T)$ of $Dy_{1-x}Tb_xNi_2B_2C$ for $H \perp c$ versus temperature (a) with $x=0.0$, 0.05, 0.1, 0.25, and 0.4 and (b) with $x=0.6$, 0.8, 0.9, and 1.0.

Figure 3 shows the susceptibilities, $\chi(T)$, for $\mathbf{H} \perp \mathbf{c}$ and $H=10$ Oe for different values of x. The data for pure $DyNi_2B_2C$ and $TbNi_2B_2C$ are also plotted for comparison. As the figure shows, the AF transitions manifest themselves with a clear deviation of $\chi(T)$, near or above $T=10$ K, from the behavior at higher T. The T_N was determined from the peak point of $\chi(T)$. For $x \leqslant 0.25$, the additional drops at lower temperatures are attributed to superconducting transitions. As x increases up to 0.4, the onset temperature of AF ordering is virtually unchanged, in spite of the increasing dG_{eff}. Instead, the transition becomes broad, and the suppression of $\chi(T)$ below T_N is weakened. As x is increased further above 0.6, the magnetic transition becomes sharper and increases, approaching that of $TbNi_2B_2C$, as seen in Fig. 3(b). This results from the facts that the Tb^{+3} ions participate more in the magnetic ordering and that the disturbance due to the Dy^{+3} ions becomes small. The upturns of $\chi(T)$ around 6 K for $x=0.8$ and 0.9 are thought to be due to the appearance of weak ferromagnetism, as in pure $TbNi_2B_2C$ around 8 K.[29]

The variations of T_c and T_N versus dG_{eff} in our Dy—Tb system, as well as in the previously reported Dy—Ho system,[18,23] are presented in Fig. 4. dG_{eff} is 7.083 for pure $DyNi_2B_2C$, and that for Dy—Tb (Dy—Ho) is larger (smaller) than this value. For Dy—Ho, the T_N scales with dG_{eff} fairly well, as the RKKY theory predicts. However, dG_{eff} scaling breaks down for T_c. The T_c is suppressed by increasing dG_{eff}, as predicted by the AG pair-breaking

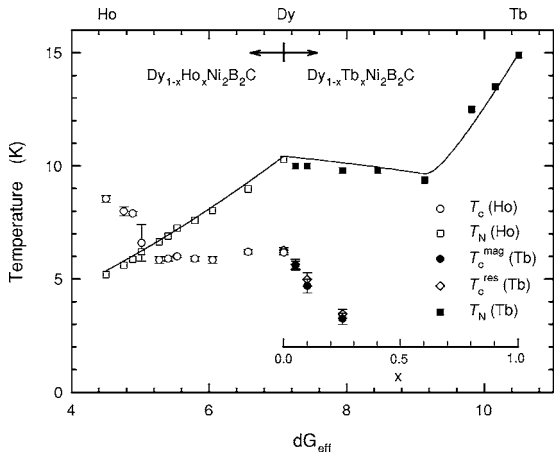

FIG. 4. Superconducting transition temperature T_c and antiferromagnetic ordering temperature T_N versus the average de Gennes factor dG_{eff} of $Dy_{1-x}Tb_xNi_2B_2C$ $[T_c(Tb_x), T_N(Tb_x)]$ and $(Dy,Ho)Ni_2B_2C$ $[T_c(Ho_x), T_N(Ho_x)]$.[16] T_c^{res} was determined from the resistance measurements while T_c^{mag} was determined from susceptibility measurements.

theory, only in the limited region where $T_c > T_N$. The breakdown of this dG_{eff} dependence suddenly shows up at the crossing point of T_N and T_c; T_c drops sharply at that point and then becomes virtually constant as long as $T_c < T_N$. This anomalous behavior is unprecedented in other magnetic superconductors.

The $Dy_{1-x}Tb_xNi_2B_2C$ compounds studied in this work showed dG_{eff} dependences for T_c and T_N that were totally different from those for Dy—Ho. Even a slight substitution in Dy—Tb suppresses the T_c severely while the T_c is robust for $x \leqslant 0.7$ ($dG_{eff} \geqslant 5.28$) in $Dy_{1-x}Ho_xNi_2B_2C$. The T_c of Dy—Tb decreases almost linearly with dG_{eff}, and its slope, dT_c/dx, a parameter for magnetic pair breaking, is found to be -11.9 ± 1.7 K. On the other hand, the T_N of Dy—Tb does not follow dG_{eff} scaling. This is also in strong contrast to the Dy—Ho case. As x increases, the T_N of Dy—Tb, ~10 K, remains almost unchanged up to $x=0.6$ ($dG_{eff}=9.13$), and then increases rapidly to the value for values up to $x=1.0$.

A phenomenological model was suggested to elucidate the pair-breaking effect of $Dy_{1-x}Ho_xNi_2B_2C$.[30] In that model, the multiband RNi_2B_2C system was assumed to have two superconducting order parameters (SOP); one was due to the Ni band, and the other was due to the other bands. In a paramagnetic state, both SOP's are suppressed as dG_{eff} increases. However, in a magnetically ordered state, the SOP from the Ni band is affected very little by the local field due to the field cancellation effect at the Ni site because geometrically, Ni is thought to be located at the center of a tetrahedron of four nearest rare-earth ions in $Dy_{1-x}Ho_xNi_2B_2C$ as in the mother compounds $DyNi_2B_2C$ and $HoNi_2B_2C$ (See the Ni ion with a cross mark in Fig. 1). In contrast, the SOP from the other bands is significantly suppressed. Therefore, the T_c is nearly constant for $T_c < T_N$. This field cancellation effect below T_N was experimentally verified in a Mössbauer study,[31] where a local pair-breaking field was observed at the Ni sites in $TbNi_2B_2C$, but not in $DyNi_2B_2C$ and $HoNi_2B_2C$. On the same basis, the suppres-

sion of the T_c in Dy—Lu by substitution of nonmagnetic ions, Lu^{+3},[18,23] can be easily understood. The substituted Lu^{+3} does not work to cancel the field caused by Dy^{+3}, but works instead as a pair breaker.

The above picture can also be applied to the Dy—Tb system. When Tb^{+3} is substituted, the R sites are occupied by the magnetic spins of Tb^{+3}, and in this case, the preferred spin direction of Tb^{+3} ([100]) is different from that of Dy^{+3} ([110]). Therefore, on Tb^{+3} substitution, because of imperfect cancellation, the electrons at the Ni site start to feel the local field due to the magnetic spins, which decreases T_c; Tb^{+3} plays the role of a magnetic pair breaker in the Dy—Tb system. This interpretation can also be confirmed by comparing the strengths of pair breaking in Dy—Tb and Dy—Lu. Since Lu^{+3} is a magnetic-spin vacancy, the remaining field at the Ni site for Lu^{+3} is estimated to be larger than that for Tb^{+3}; therefore, in $DyNi_2B_2C$, a Lu^{+3} ion will act as a stronger pair breaker than a Tb^{+3} ion. As expected, the T_c suppression rate for Lu^{+3} substitution, $dT_c/dx \approx -37$ K, is much larger than that for Tb^{+3} substitution, $dT_c/dx \approx -12$ K.

The T_N of the Dy—Tb system does not follow de Gennes scaling, but this breakdown of de Gennes scaling is expected because $DyNi_2B_2C$ and $TbNi_2B_2C$ have different magnetic ordering structure. The wave vector of the magnetic order of $DyNi_2B_2C$ is $Q=(0,0,2\pi)$ in the magnetic ground state, but that of $TbNi_2B_2C$ is $Q \approx (1.1\pi,0,0)$. These ground states of the two mother compounds will compete with each other in the Dy—Tb system. This competition or the change of magnetic structures can result in a breakdown of de Gennes scaling in T_N. Possibly, for the Dy-rich side, the substitution of Tb increases dG, but randomizes the magnetic structure, which compensates for the increase in dG. Therefore, T_N does not change. In contrast, for the Tb-rich side, the substitution of Tb causes the overall magnetic structure to resemble that of $TbNi_2B_2C$. Thus, the detailed magnetic structures of $Dy_{1-x}Tb_xNi_2B_2C$ as a function of x must be determined, for example, by using neutron scattering experiments.

The above-mentioned scenario could also explain a nonmonotonic behavior of T_N in $Er_{1-x}Tb_xNi_2B_2C$ polycrystalline samples.[32,33] In this Er—Tb system, T_N remained almost constant for concentrations up to $x=0.5$ but beyond this value, T_N increased rapidly. This behavior is very similar to that of the Dy—Tb system and therefore, the competition of magnetic ground states of the two mother compounds is also thought to play an important role in determining T_N in the Er—Tb system.

IV. CONCLUSION

We studied the superconductivity and the magnetism of a series of $Dy_{1-x}Tb_xNi_2B_2C$ single crystals for which the magnetic structures of the mother compounds were different from each other. The $T_c(x)$ is suppressed rapidly by Tb substitution, but the $T_N(x)$ is almost constant for $x \leqslant 0.6$. This is in strong contrast to the $Dy_{1-x}Ho_xNi_2B_2C$ case where the mother compounds have similar magnetic structures; the $T_c(x)$ of Dy—Ho is virtually constant for $x<0.7$, i.e., $T_c<T_N$, and the $T_N(x)$ follows de Gennes scaling well.

After comparing the three cases of R' in $Dy_{1-x}R'_xNi_2B_2C$, a nonmagnetic ion (Lu^{+3}), a magnetic ion with similar spin anisotropy and magnetic ordering (Ho^{+3}), and a magnetic ion with different anisotropy and magnetic ordering (Tb^{+3}), we were able to present an explanation that could account for the superconductivities and the magnetisms of all three cases. The pair breaking or the T_c depression in $Dy_{1-x}R'_xNi_2B_2C$ for $T_c<T_N$ mainly originates from the uncanceled field at the Ni site as in Dy—Tb or Dy—Lu. In the determination of the T_N of a pseudoquaternary $R_{1-x}R'_xNi_2B_2C$, not only the de Gennes factor but also the magnetic ordering are crucial. When R and R' elements having distinct magnetic ordering states in RNi_2B_2C, like Dy and Tb, are mixed, the T_N of $R_{1-x}R'_xNi_2B_2C$ cannot be estimated from only the change in the dG_{eff}.

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