

## Effects of Tm substitution on superconductivity and magnetism in the antiferromagnetic borocarbide superconductor $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$

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(Received 29 January 2010; revised manuscript received 26 April 2010; published 18 June 2010)

A series of single-phased  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) compounds were prepared by an arc-melting method. The superconductivity and magnetic properties have been investigated by measuring electrical resistivity, magnetization, and specific heat. The superconducting transition temperature  $T_c$  decreases rapidly with increasing  $x$ , and shows a minimum around  $x=0.15$ , then increases gradually with a further increase in  $x$ . The magnetic transition temperature  $T_M$  gradually decreases with increasing  $x$ . The effective magnetic moment  $\mu_{\text{eff}}$  gradually decreases with increasing  $x$  and agrees with the estimation assuming the free ion values of  $\text{Dy}^{3+}$  and  $\text{Tm}^{3+}$  states, indicating that the change in the electronic structure of Dy and Tm ions is very small. The present results together with previously reported  $\text{Dy}_{1-x}\text{R}_x\text{Ni}_2\text{B}_2\text{C}$  ( $R=\text{Ho}, \text{Tb}, \text{Y}, \text{and Lu}$ ) systems were discussed and well explained in the frame of Abrikosov and Gor'kov theory and the field cancellation effect at Ni sites.

DOI: 10.1103/PhysRevB.81.214517

PACS number(s): 74.25.Dw, 74.62.Dh

### I. INTRODUCTION

Since the discovery of quaternary nickel boroncarbides  $\text{RNi}_2\text{B}_2\text{C}$  ( $R=\text{Y}$  or rare earth) in January 1994,<sup>1-3</sup> these compounds have attracted considerable attention by several groups. The compounds crystallize in the tetragonal  $\text{ThCr}_2\text{Si}_2$ -like structure, which can be displayed as a frame with alternating  $R\text{-C}$  and  $\text{Ni}_2\text{B}_2$  layers. Despite this layered structure, band-structure calculations have shown a three-dimensional electronic behavior.<sup>4,5</sup> The compounds have a multiband character, and the  $3d$  electrons at Ni are major charge carriers and mainly contribute to superconductivity, even though the contributions from other bands are not ignorable. According to previous reported results,<sup>1-16</sup> the Pr, Nd, Dy, and Ho systems have commensurate antiferromagnetic structures, and the Gd, Tb, Er, and Tm systems form incommensurate magnetic structures. Among these compounds, the Y, Lu, Dy, Ho, Er, and Tm systems exhibit superconductivity. For the Dy, Ho, Er, and Tm systems, superconductivity coexists with magnetic order; the ratio of superconducting transition temperature ( $T_c$ ) to antiferromagnetic ordering temperature  $T_N$  ranges from  $T_c/T_N=7.0$  for Tm to 0.60 for Dy systems.

Early studies suggest that at least for the quaternary parent compounds,  $T_N$  as well as  $T_c$  can be well scaled with the de Gennes (dG) factor  $(g_J-1)^2J(J+1)$ , where  $g_J$  is the Landé  $g$  factor and  $J$  is the total angular moment of the  $R^{3+}$  ion estimated for the Hund's rule ground state.<sup>9-12</sup> This scaling of  $T_N$  can be understood in terms of the conduction-electron-mediated Ruderman-Kittel-Kasuya-Yosida coupling between rare-earth ions, which is an exchange interaction between localized magnetic spins and conduction electrons.<sup>14,15</sup> The linear dependence of  $T_c$  on the dG factor is consistent with the predictions of the Abrikosov-Gor'kov (AG) theory<sup>17</sup> of the pair-breaking effect by magnetic impurities. According to the AG theory,  $T_c$  is rapidly suppressed with the increasing of dG. In pure  $\text{RNi}_2\text{B}_2\text{C}$ , in which the magnetic elements  $R$  are located on regular lattice sites,  $T_c$  decreases monotonically with the increasing of dG from  $R=\text{Y(Lu)}$  to  $R=\text{Dy}$ .<sup>14</sup> Also,

for mixed  $\text{R}_{1-x}\text{R}'_x\text{Ni}_2\text{B}_2\text{C}$  systems, superconductivity is suppressed as the effective dG ( $dG_{\text{eff}}$ ) factor,  $dG_{\text{eff}}=(1-x) \times dG[R]+x \times dG[R']$ , increases as long as the  $T_c$  is higher than  $T_N$  as predicted by the AG theory. However, it has been reported that for several pseudoquaternary  $\text{R}_{1-x}\text{R}'_x\text{Ni}_2\text{B}_2\text{C}$  systems the de Gennes scaling of  $T_c$  or/and  $T_N$  breaks down entirely.<sup>16,18-23</sup> It should be noted that in the case of  $R=\text{Dy}$ , superconductivity appears in the magnetic ordered state and in this case, AG theory still valid or not is an open question.<sup>14,19,20</sup> Very recently, a reversible giant magnetocaloric effect were observed in  $\text{Dy}_{0.9}\text{Tm}_{0.1}\text{Ni}_2\text{B}_2\text{C}$ .<sup>24</sup> The magnetic phase transition  $T_M$  and superconducting transition temperature  $T_c$  are 9.2 K and 4.5 K in  $\text{Dy}_{0.9}\text{Tm}_{0.1}\text{Ni}_2\text{B}_2\text{C}$ , respectively, lower than those in  $\text{DyNi}_2\text{B}_2\text{C}$ .<sup>24</sup> This behavior was similar to that of nonmagnetic substituted  $\text{Dy}_{1-x}\text{Lu}_x\text{Ni}_2\text{B}_2\text{C}$  system.<sup>16,22</sup> Thus, Tm substitution to the Dy site in  $\text{DyNi}_2\text{B}_2\text{C}$  system may be another typical candidate to study the coexistence and competition of magnetism and superconductivity. To further understand these phenomena clearly, in this paper we report a systematic study of superconductivity, magnetic, and thermodynamic properties in  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) system, and the phase diagram is determined.

### II. EXPERIMENTAL PROCEDURE

Polycrystalline samples of  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) were prepared by an arc-melting method using a tungsten electrode under an argon atmosphere. First, we melted the stoichiometric amounts of Dy, Tm, Ni, B, and C on a water-cooled copper hearth. The weight loss of the sample was attributed to boron and was accordingly compensated. Then, the sample was melted more than six times for homogeneity. The total weight loss of the sample obtained by this method is less than 0.5%. Then the samples were finally annealed at 1323 K for 72 h in evacuated quartz tubes. All the samples have the same  $\text{LuNi}_2\text{B}_2\text{C}$ -type structure with a space group of  $I/4mmm$  which was confirmed by x-ray powder-diffraction experiment. The samples were cut into rectangu-

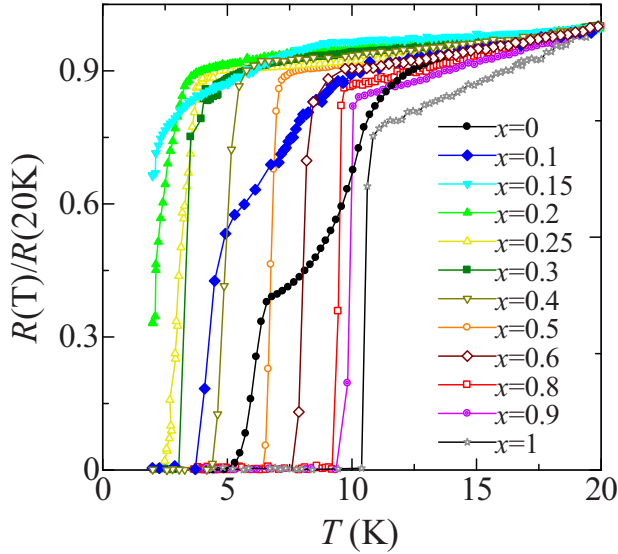


FIG. 1. (Color online) Temperature dependence of normalized electrical resistivity  $\rho(T)/\rho(20)$  for  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) system from 2 to 20 K.

lar pieces for measurements of electrical resistivity, which was carried out using a standard four-probe technique in the temperature range from 1.8 to 20 K. Magnetization measurement were carried out using a superconducting quantum interference device (Quantum Design magnetic property measurement system) in the temperature range from 2 to 300 K. Specific heat measurements were carried out by the adiabatic heat relaxation method in the temperature range from 2 to 30 K using the physical property measurement system (Quantum Design).

### III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of normalized electrical resistivity  $\rho(T)/\rho(20)$  for  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) system from 2 to 20 K. For the samples with  $x=0.15$  and  $0.20$ , the presently studied temperature seems not sufficiently low enough to make the resistivity zero but the observed significant drop of resistivity in the low-temperature region is most likely due to superconductivity. Figure 2 shows the temperature dependence of low-field dc magnetization ( $H=3$  mT) for  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) system from 2 to 20 K. For the superconducting samples, the low-field dc magnetization becoming negative or a significant drop was found near  $T_c$ . The clear  $\lambda$  shape behavior for some samples show the antiferromagnetic transition. A clear change in the slope of  $\rho(T)$  curves (Fig. 1) was also observed in the vicinity of the magnetic transition temperature. Figure 3 shows the temperature dependence of the specific heat  $C$  for  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) system. The large peaks of  $C$  are ascribed to the contributions from the magnetic transitions. The peak heights decrease and their position shift toward lower  $T$  with increasing  $x$ . The contributions of the superconducting transitions to  $C$  seem too small to be seen except for  $x=0.9$  and  $1.0$ . The superconducting transition temperature  $T_c$  and magnetic transition temperature  $T_N$  as a function of  $x$  and the  $dG_{\text{eff}}$  factor (which was calculated

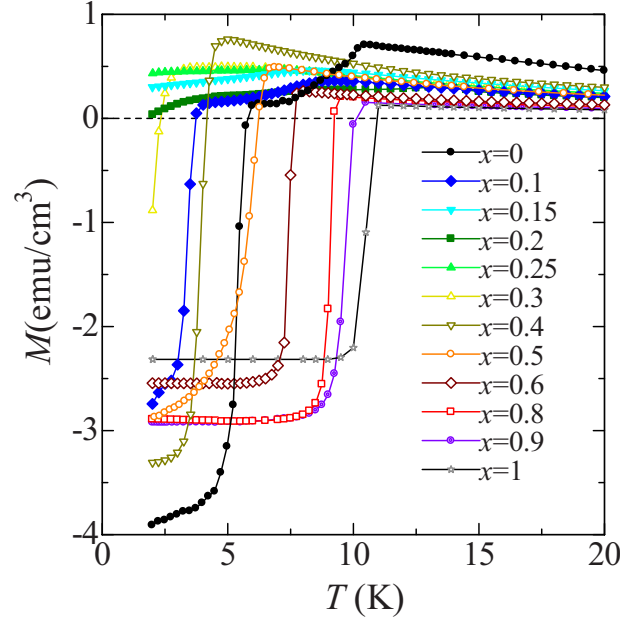


FIG. 2. (Color online) Temperature dependence of low-field dc magnetization ( $H=3$  mT) for  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) system from 2 to 20 K.

using  $dG_{\text{eff}}=(1-x) \times dG[\text{Dy}]+x \times dG[\text{Tm}]$  for  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) system are shown in Fig. 4. We can note that  $T_c$  decreases rapidly with increasing  $x$  and shows a minimum around  $x=0.15$  ( $dG$  factor  $\sim 6.2$ ), then increases gradually with a further increase in  $x$ , i.e., a totally break down of  $dG$  scaling. The magnetic transition temperature  $T_N$  deduced from  $C(T)$  curves was consistent with that from low-field  $M(T)$  curves, and  $T_N$  gradually decreases with decreasing  $dG$  factor (increases in  $x$ ), i.e., it can be well scaled by the  $dG$  factor. Moreover, there is no obvious change at the cross point of  $T_c$  and  $T_N$ .

The temperature dependence of the reciprocal susceptibility  $1/\chi$  at an external field of 1 T for  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) system is shown in Fig. 5. The reciprocal susceptibility in high-temperature region could be well described by the Curie-Weiss law, i.e.,

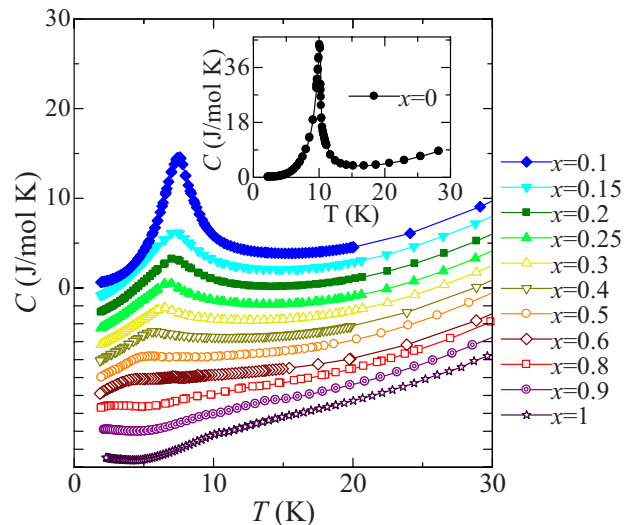


FIG. 3. (Color online) Temperature dependence of the specific heat  $C$  for  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) system.

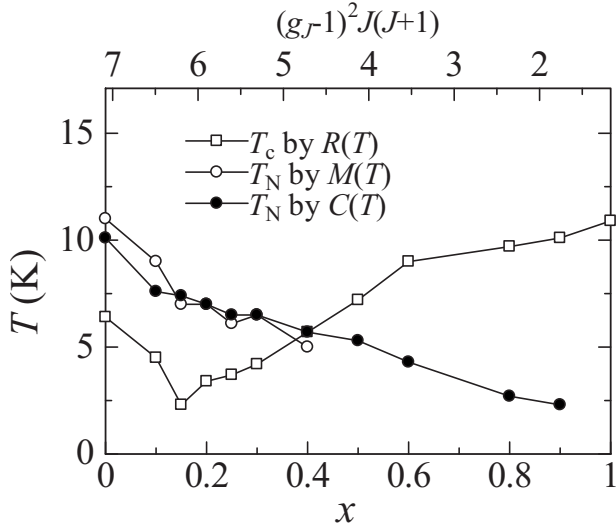


FIG. 4. Superconducting transition temperature  $T_c$  and magnetic transition temperature  $T_N$  as functions of  $x$  and  $dG_{\text{eff}}$  factor (which was calculated using  $dG_{\text{eff}}=(1-x) \times dG[\text{Dy}] + x \times dG[\text{Tm}]$ ) for  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) system.

$$\chi = C/(T - \theta_p), \quad (1)$$

where  $C$  is the Curie constant and  $\theta_p$  is the paramagnetic Curie constant. The effective magnetic moment  $\mu_{\text{eff}}$  deduced from the Curie-Weiss law as a function of  $x$  in  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  system is shown in Fig. 6. The value of  $\mu_{\text{eff}}$  gradually decreases with increasing  $x$ . The estimated values of  $\mu_{\text{eff}}$  for the  $\text{DyNi}_2\text{B}_2\text{C}$  and  $\text{TmNi}_2\text{B}_2\text{C}$  are  $10.29\mu_B$  and  $7.33\mu_B$ , which are close to the values of free ions;  $\text{Dy}^{3+}$  ( $10.63\mu_B$ ) and  $\text{Tm}^{3+}$  ( $7.55\mu_B$ ), respectively. Indeed, the experimental derived values of  $\mu_{\text{eff}}$  are consistent with those of theoretical calculated (as shown in Fig. 6) by the following equation:<sup>25</sup>

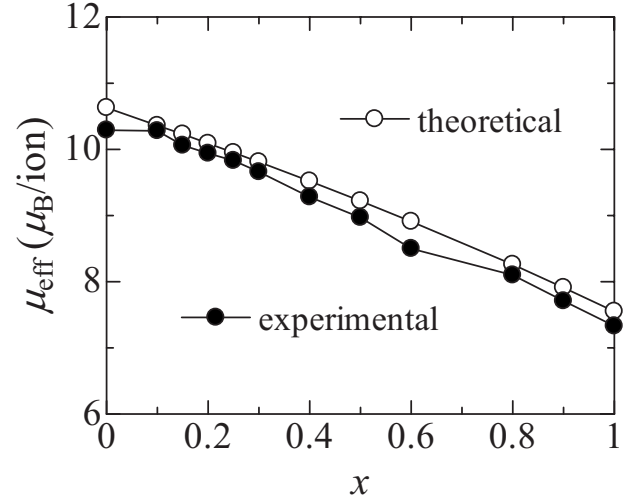


FIG. 6. The values of experimentally deduced (closed symbols) and theoretically calculated (open symbols) effective magnetic moment  $\mu_{\text{eff}}$  as a function of  $x$  in  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  system.

$$[\mu_{\text{eff}}(\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C})]^2 = x \times [\mu_{\text{eff}}(\text{Dy}^{3+})]^2 + (1-x) \times [\mu_{\text{eff}}(\text{Tm}^{3+})]^2. \quad (2)$$

This results indicates that the electronic structures of  $\text{Dy}^{3+}$  and  $\text{Tm}^{3+}$  ions in the ground state do not have a pronounce change in the entire  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  system. The temperature dependence of high-field magnetic susceptibility at a field of 1 T (higher than  $H_{c2}$ ) for  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) system is also shown in the inset of Fig. 5. The magnetic susceptibility shows a weak temperature dependence at low temperatures for  $x=0.2-0.6$ , i.e., a ferrimagneticlike behavior emerged. The magnetic field dependence of magnetization for  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) system at 2 K up to 7 T was measured, and the results are shown in Fig. 7. The results of samples for  $x=0$  and 0.1 are consistent with those of previously reported by Li and Nishimura.<sup>24</sup> The magneti-

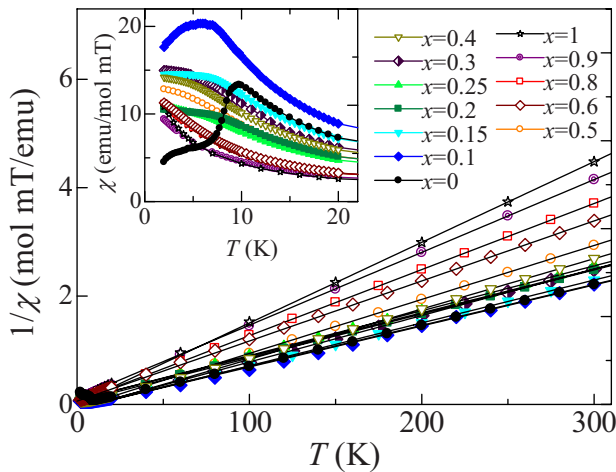


FIG. 5. (Color online) Temperature dependence of the reciprocal susceptibility  $1/\chi$  at an external field of 1 T for  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) system. Inset shows the temperature dependence of magnetic susceptibility at a field of 1 T (higher than  $H_{c2}$ ) for  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) system.

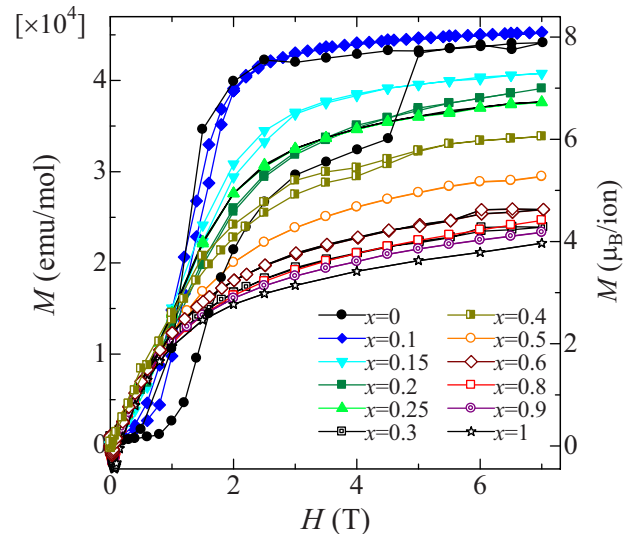


FIG. 7. (Color online) Magnetic field dependence of magnetization for  $\text{Dy}_{1-x}\text{Tm}_x\text{Ni}_2\text{B}_2\text{C}$  ( $x=0-1$ ) system at 2 K up to 7 T with increasing and decreasing field.

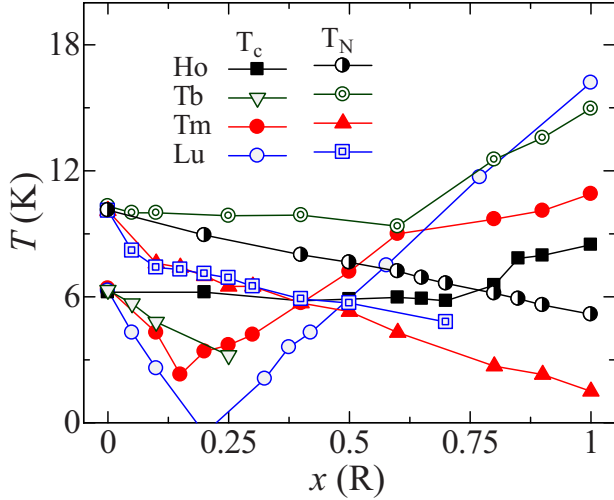


FIG. 8. (Color online) Superconducting transition temperature  $T_c$  and magnetic transition temperature  $T_N$  as functions of  $x$  in  $\text{Dy}_{1-x}\text{R}_x\text{Ni}_2\text{B}_2\text{C}$  ( $R=\text{Ho}$ ,  $\text{Tb}$ ,  $\text{Tm}$ , and  $\text{Lu}$ ;  $x=0-1$ ) system.

zation tends to be saturated at high field which is similar to those in the Y or Lu substituted  $\text{DyNi}_2\text{B}_2\text{C}$  system.<sup>20,21,26</sup> Morozov<sup>27</sup> theoretically predicated that similarly as in the spin-triplet paired superconductors, nonmagnetic impurity in an antiferromagnetic superconductor cause pair breaking. Morozov<sup>28</sup> also theoretically studied the reentrant behavior of  $\text{HoNi}_2\text{B}_2\text{C}$  in the temperature region  $5 < T < 6$  K, and concluded that the modification of the wave functions of conduction electrons by the long-range magnetic order due to the paramagnetic phase is the main reason for the abrupt suppression of superconductivity. Nass *et al.*<sup>29</sup> also theoretically studied the impurity effect on superconductivity. They concluded that in some magnetic superconductors, nonmagnetic impurities may suppress  $T_c$  owing to the destruction of the translational symmetry of the antiferromagnetic lattice. In the present Tm-substituted system and our previously reported Lu-substituted  $\text{DyNi}_2\text{B}_2\text{C}$  system,<sup>16</sup> the observed ferrimagneticlike magnetic order at a certain substitution content can be a main reason for the suppression of superconductivity. The observed ferrimagneticlike behavior is possibly due to some spin fluctuation caused by the disorder and inhomogeneity that was induced by Tm substitution. These behaviors were similar to  $\text{Dy}_{1-x}\text{Lu}_x\text{Ni}_2\text{B}_2\text{C}$  system.<sup>16</sup>

It has been also demonstrated that for pseudoquaternary  $\text{Dy}_{1-x}\text{R}_x\text{Ni}_2\text{B}_2\text{C}$  ( $R=\text{Ho}$ ,  $\text{Tb}$ ,  $\text{Y}$ , and  $\text{Lu}$ ) systems the  $dG$  scaling of  $T_c$  or/and  $T_N$  breaks down entirely. For example, in the  $\text{Dy}_{1-x}\text{Ho}_x\text{Ni}_2\text{B}_2\text{C}$  system,<sup>18</sup>  $T_c$  was almost constant in the region of  $T_c < T_N$ . An almost unchanged  $T_N$  was found in the  $\text{Dy}_{1-x}\text{Tb}_x\text{Ni}_2\text{B}_2\text{C}$  system for  $x < 0.6$ .<sup>19</sup> The rapid suppression of superconductivity in nonmagnetic Y or Lu diluted  $\text{DyNi}_2\text{B}_2\text{C}$  systems was also observed.<sup>16,20-23</sup> To further understand the Dy site substitution effect, the variations in  $T_c$  and  $T_N$  versus doping content  $x$  and  $dG_{\text{eff}}$  factor in  $\text{Dy}_{1-x}\text{R}_x\text{Ni}_2\text{B}_2\text{C}$  ( $R=\text{Ho}$ ,  $\text{Tb}$ ,  $\text{Tm}$ , and  $\text{Lu}$ ) system are presented in Figs. 8 and 9, respectively.

To thoroughly understand the change in  $T_c$ , we divide the substitution range into two regions where superconductivity show a minimum ( $x \sim 0.15$ ,  $0.15$ ,  $0.4$ , and  $0.7$  for  $R=\text{Lu}$ ,  $\text{Tm}$ ,  $\text{Tb}$ , and  $\text{Ho}$ , respectively) and discuss them separately. From

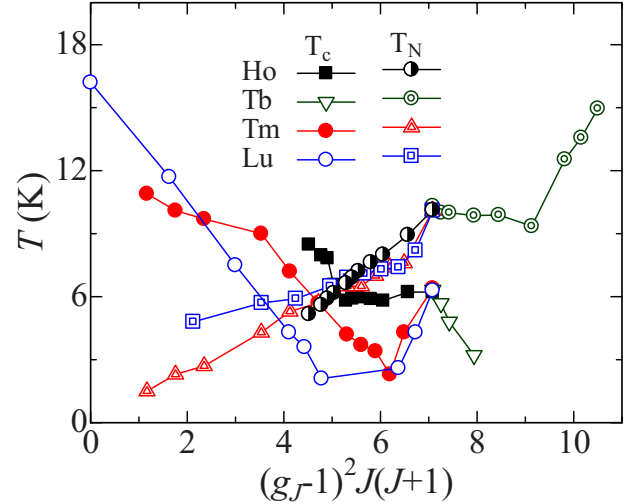


FIG. 9. (Color online) Superconducting transition temperature  $T_c$  and magnetic transition temperature  $T_N$  as functions of  $dG_{\text{eff}}$  factor (which was calculated using  $dG_{\text{eff}}=(1-x) \times dG[\text{Dy}] + x \times dG[\text{Tm}]$ ) in  $\text{Dy}_{1-x}\text{R}_x\text{Ni}_2\text{B}_2\text{C}$  ( $R=\text{Ho}$ ,  $\text{Tb}$ ,  $\text{Tm}$ , and  $\text{Lu}$ ) system.

Fig. 8, for the low doping content,  $T_c$  almost linearly decreases with increasing  $R$  content  $x$ , and the suppression rate of  $T_c$  on  $x$ ,  $\partial T_c / \partial x$ , is  $-37$ ,  $-26$ ,  $-12$ , and  $\sim 0$  K for  $R=\text{Lu}$ ,  $\text{Tm}$ ,  $\text{Tb}$ , and  $\text{Ho}$ , respectively. According to the AG theory,<sup>17</sup> the magnetic pair-breaking effect is due to the exchange scattering by uncorrelated local magnetic spins, and the suppression of superconductivity can be characterized by

$$\partial T_c / \partial x \propto -J_{\text{sf}}^2 \times dG, \quad (3)$$

where  $x$  is the concentration of magnetic moment and  $J_{\text{sf}}$  is an exchange coupling constant between the local moments and the conduction electrons. From Fig. 9, for  $\text{Dy}_{1-x}\text{R}_x\text{Ni}_2\text{B}_2\text{C}$  system at low doping content (corresponding  $dG_{\text{eff}}$  in the range of 6–8 in Fig. 9),  $T_c$  decreases with increasing  $dG_{\text{eff}}$  for  $R=\text{Tb}$  as is predicated by AG theory. However,  $T_c$  increases with the increases in  $dG_{\text{eff}}$  for  $R=\text{Lu}$  and  $\text{Tm}$ , which is contradict with AG theory. The almost unchanged in  $T_c$  for  $R=\text{Ho}$  also cannot be understood with this theory. Doh *et al.*<sup>30</sup> proposed a phenomenological model which includes two magnetic and two superconducting order parameters (SOPs) accounting for the multiband structure in  $\text{RNi}_2\text{B}_2\text{C}$  system, and the pair-breaking effect in  $\text{Dy}_{1-x}\text{Ho}_x\text{Ni}_2\text{B}_2\text{C}$  system was well explained. Based on this model, the dominant one of the SOPs was due to the Ni band, and the other SOP was due to the other bands. In a paramagnetic state, both SOPs are suppressed as  $dG_{\text{eff}}$  increases. However, in a magnetically ordered state, the SOP from the Ni band is affected 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 the tetrahedron of four nearest rare-earth ions in the  $\text{Dy}_{1-x}\text{R}_x\text{Ni}_2\text{B}_2\text{C}$  system. The band originating from Ni does not feel the magnetic moment of Dy(Ho) anymore below the Neel temperature since it is located exactly in the center of a tetrahedron of the nearest Dy(Ho) atoms. This is the reason why  $T_c$  almost unchanged below  $T_N$ . This behavior was also experimentally verified in a Mossbauer study.<sup>31</sup> Therefore, with the substitution of



other  $R^{3+}$  at  $Dy^{3+}$  site, the field cancellation effect will modify the wave functions of conduction electrons by the long-range magnetic order in  $Dy_{1-x}R_xNi_2B_2C$  system.<sup>4,5</sup> The observed ferrimagneticlike magnetic order in Tm- and Lu-substituted  $DyNi_2B_2C$  systems properly reflected the field cancellation effect by the random doping at Dy site. The strength can be simply estimated from the difference in magnetic properties, including the magnetic moment and the magnetic ordering wave vector, of  $RNi_2B_2C$  from the mother compound  $DyNi_2B_2C$ . The AG theory is still valid as well if we consider the above points and assume the difference in magnetic properties as  $|\Delta dG_{\text{eff}}|$ , i.e., for a magnetic ordered superconductor, Eq. (3) can be modified as

$$\partial T_c / \partial x \propto -I_{\text{sf}}^2 \times |\Delta dG_{\text{eff}}| - J_{\text{sf}}^2 \times dG_{\text{eff}} \quad (4)$$

in which  $I_{\text{sf}}$  is the exchange coupling constant between the Ni(3d) electrons and the effective magnetic cancellation field. The magnetic structure of  $DyNi_2B_2C$  and  $HoNi_2B_2C$  consist of ferromagnetically aligned Dy(Ho) spins in the  $ab$  plane but antiferromagnetically coupled along the  $c$  axis.<sup>14,20</sup> For the  $TbNi_2B_2C$ , the magnetic spins points along the  $a$  axis and form an  $a$ -axis-modulated order structure.<sup>8</sup> The antiferromagnetic ground state of  $TmNi_2B_2C$  consists of ferromagnetic planes along the  $c$  axis, sinusoidally modulated along the (110) direction.<sup>32</sup> The values of dG factor are 1.17, 4.5, 7.1, and 10.5 for  $Tm^{3+}$ ,  $Ho^{3+}$ ,  $Dy^{3+}$ , and  $Tb^{3+}$ , respectively, and  $T_N$  increases in this order. For the nonmagnetic  $Lu^{3+}$  is a magnetic spin vacancy, the values of dG factor will be 0, therefore,  $Lu^{3+}$  ion will act as the stronger pair breaker than the other magnetic ordered  $R^{3+}$  ions. As a matter of fact, the suppression rate  $\partial T_c / \partial x$  for  $R=Lu, Tm, Tb$ , and  $Ho$  is well consistent with the predicted. From Fig. 9, at low doping content ( $dG_{\text{eff}}$  in the range of 6–8), we can note that the absolute value of suppression rate of  $T_c$  on  $dG_{\text{eff}}$ ,  $|\partial T_c / \partial dG_{\text{eff}}|$ , does not show too much difference for  $R=Lu, Tm$ , and  $Tb$ . That is, the pair-breaking effect in  $DyNi_2B_2C$  mainly depends on the field cancellation effect at the Ni site, it is not directly related to the magnetic moment of the dopant ions at least for  $R=Lu, Tm$ , and  $Tb$  in  $Dy_{1-x}R_xNi_2B_2C$  system.

For the higher doping region ( $x > 0.3, 0.15$ , and  $0.75$  for  $R=Lu, Tm$ , and  $Ho$ , respectively), the behavior of superconductivity is easily understood. The decreases in  $T_c$  with increasing Dy content ( $1-x$ ) is due to the magnetic pair-breaking effect when the rare-earth ions are replaced by the magnetic Dy ions for  $RNi_2B_2C$  ( $R=Lu, Tm$ , and  $Ho$ ), as predicted by the AG theory.<sup>14,17</sup> However, the  $\partial T_c / \partial dG_{\text{eff}}$  versus dG factor for these compounds (in Fig. 9) seems not to show a universal behavior which was different from the AG theory prediction. This behavior originated from different crystalline electric field (CEF) effects of  $R^{3+}$  and different values of conduction-electron-local-moment coupling constant.<sup>9,22</sup> From Fig. 9, in the range of  $dG_{\text{eff}} \sim 3-6$  and  $4.5-5$  for  $R=Tm$  and  $Ho$ , respectively, the value of  $\partial T_c / \partial dG_{\text{eff}}$  for  $R=Tm$  and  $Ho$  is almost the same as  $R=Lu$  ( $dG_{\text{eff}} \sim 0-5$ ) which possibly suggests that CEF of  $Dy^{3+}$  plays dominant role at these regions.

From Figs. 8 and 9, we can note that both  $x$  and  $dG_{\text{eff}}$  dependence of  $T_N$  in  $Dy_{1-x}Tm_xNi_2B_2C$  show a linear behavior. However, the  $T_N$  does not follow the dG scaling in  $Dy_{1-x}Tb_xNi_2B_2C$  (Ref. 19) and  $Er_{1-x}Tb_xNi_2B_2C$  (Ref. 33) systems because the two mother compounds have different magnetic ordering structure. These ground states of the two mother compounds will compete with each other and this competition or the change in magnetic structures can result in a breakdown of dG scaling. The present  $TmNi_2B_2C$  and  $DyNi_2B_2C$  have different magnetic structures but the  $T_N$  of  $TmNi_2B_2C$  (1.5 K) is much lower than that in  $DyNi_2B_2C$  (10.6 K), i.e., the magnetic exchange interactions of Tm moments are smaller than those of Dy moments. The breakdown of dG scaling in  $Dy_{1-x}Tm_xNi_2B_2C$ , therefore, is expected to be observed at very small amount of Dy content region (no data in present study). The linear dependence on  $x$  and  $dG_{\text{eff}}$  of  $T_N$  in  $Dy_{1-x}R_xNi_2B_2C$  ( $R=Lu$  and  $Ho$ ) can reflect the same magnetic structure in the whole doping range in these compounds.

#### IV. CONCLUSIONS

In summary, we have systematically studied the superconductivity and magnetism of  $Dy_{1-x}Tm_xNi_2B_2C$  ( $x=0-1$ ) compounds. For lower doping region ( $x < 0.2$ ), the superconducting transition temperature  $T_c$  decreases rapidly with increasing Tm content  $x$  and shows a minimum around  $x=0.15$ , which is mainly due to the field cancel effect at Ni sites. For the higher doping region ( $x \geq 0.2$ ),  $T_c$  decreases gradually with a further increases in Dy content  $1-x$  (increases with Tm content  $x$ ), which is due to the magnetic pair-breaking effect when the Tm ions are replaced by the magnetic Dy ions. The magnetic transition temperature  $T_M$  and the effective magnetic moment gradually decreases with increases  $x$ . From a comparison study of  $x$  and  $dG_{\text{eff}}$  dependences of  $T_c$  and  $T_N$  in  $Dy_{1-x}R_xNi_2B_2C$  ( $R=Ho, Tb, Tm$ , and  $Lu$ ) systems, based on the AG theory<sup>17</sup> and the phenomenological model proposed by Doh *et al.*,<sup>30</sup> we presented an explanation that could account for the superconductivity and the magnetism of the magnetic ordered superconductors  $Dy_{1-x}R_xNi_2B_2C$  systems. We can conclude that the pair-breaking effect in  $DyNi_2B_2C$  does not directly relate to the magnetic moment of the dopant ions, it mainly depends on the field cancellation effect at the Ni site at least for  $Dy_{1-x}R_xNi_2B_2C$  ( $R=Lu, Tm$ , and  $Tb$ ) systems. The relation between  $T_N$  and  $dG_{\text{eff}}$  (following or breaking dG scaling) mainly depends on the magnetic structure in the ground state of the mother compound.

#### ACKNOWLEDGMENTS

This work was partially supported by the National Natural Science Foundation of China (Grant No. 50871036), Innovation Research Team for Spintronic Materials and Devices of Zhejiang Province, and Major Scientific and Technological Innovation Project for Higher Education of Zhejiang Province (Grant No. ZD2007010).

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