Phenomenological Theory of Superconductivity and Magnetism in $Ho_{1-x}Dy_xNi_2B_2C$

Hyeonjin Doh,[1] Manfred Sigrist,[2] B. K. Cho,[1,*] and Sung-Ik Lee[1]
[1]*National Creative Research Initiative Center for Superconductivity and Department of Physics, Pohang University of Science and Technology, Pohang 790-784, Korea*
[2]*Yukawa Institute for Theoretical Physics, Kyoto University, Kyoto 606-8502, Japan*
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The coexistence of the superconductivity and magnetism in the $Ho_{1-x}Dy_xNi_2B_2C$ is studied by using Ginzburg-Laudau theory. This alloy shows the coexistence and complex interplay of superconducting and magnetic order. We propose a phenomenological model which includes two magnetic and two superconducting order parameters accounting for the multiband structure of this material. We describe phenomenologically the magnetic fluctuations and order and demonstrate that they lead to anomalous behavior of the upper critical field. The doping dependence of T_c in $Ho_{1-x}Dy_xNi_2B_2C$ showing a reentrance behavior is analyzed yielding very good agreement with experimental data.

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The discovery of the series of nickelborocarbide, RNi_2B_2C (R = Y, Yb, Dy, Ho, Er, Tm, and Tb) [1,2], a few years ago opened a new chapter in the discussion of magnetic superconductors [3]. This class of materials shows variety of magnetic ordered states [4] depending on the rare-earth atoms R, because the magnetism is due to the spins of their localized $4f$ orbital coupled via RKKY interaction. Some of these compounds show superconductivity with a comparatively high transition temperature ($T_c = 16.6$ K for $LuNi_2B_2C$). Band calculations of nonmagnetic $LuNi_2B_2C$ [5,6] reveal a complicated multiband structure with a rather high density of states at the Fermi level. Superconductivity is likely to be conventional originating from electron-phonon interaction due to the easily polarizable light ions B and C. The relatively large transition temperature is then a consequence of the enhanced density of states.

An interesting aspect of this material class is that some members (R = Tm, Er, Ho, and Dy) exhibit simultaneously both magnetism and superconductivity. They have a rather wide range of the ratio of T_c/T_N from 0.6 for R = Dy to 7.0 for R = Tm. Among them, $HoNi_2B_2C$ and $DyNi_2B_2C$ constitute ideal materials to study the relation between superconductivity and magnetism. They display the same antiferromagnetic order, but have different T_N due to a different de Gennes factor (dGF), $(g_J - 1)^2 \times J(J + 1)$. This leads to the situation that the superconducting T_c lies above (below) T_N for the Ho (Dy) compound. Therefore, it is natural to investigate how the two phases develop in the alloy $Ho_{1-x}Dy_xNi_2B_2C$ [7]. The experiments show that there is no simple de Gennes scaling of T_c which is expected to monotonically decrease with increasing $(g_J - 1)^2 J(J + 1)$ according to the Abrikosov-Gor'kov theory, if we assume the R ions enter as magnetic impurities. This is not the case here, since the R ions form a regular lattice with magnetic order. In the crossing region of T_N and T_c the superconducting phase boundary as a function of doping concentration x is apparently discontinuous as a consequence of a reentrant behavior [7]. The purpose of this Letter is to explain these and further experimental features consistently within a single phenomenological model including both magnetism and superconductivity.

The neutron scattering and magnetization experiments of $HoNi_2B_2C$ [8] and $DyNi_2B_2C$ show that the magnetic properties of both materials are highly anisotropic with an easy axis in the [110] direction [4,9–11]. Both $HoNi_2B_2C$ and $DyNi_2B_2C$ have antiferromagnetic (AF) order below $T_N = 5$ and 10 K, respectively. The spins order ferromagnetically within the a-b plane and antiferromagnetically in the c direction. For $HoNi_2B_2C$, there exists a significant spiral (SP) short-range correlation with the wave vector, $\mathbf{Q}^* \simeq 0.91\mathbf{Q}$, where $\mathbf{Q}$ is the AF wave vector; i.e., the spins are aligned ferromagnetically in the a-b plane, and rotate by $\phi \sim 163°$ between adjacent planes. A plausible microscopic origin of these two magnetic correlations has been discussed by Kalatsky *et al.* [12] and Amici *et al.* [13].

Based on this experimental information, we construct our phenomenological model of two magnetic order parameters, M_{AF} and M_{SP}, corresponding to the two dominant correlations. The effective Ginzburg-Landau free energy has the form

$$\mathcal{F}_M = \alpha_{\mathrm{AF}}(T){M_{\mathrm{AF}}}^2 + \alpha_{\mathrm{SP}}(T){M_{\mathrm{SP}}}^2 + \frac{\beta_{\mathrm{AF}}}{2}{M_{\mathrm{AF}}}^4 + \frac{\beta_{\mathrm{SP}}}{2}{M_{\mathrm{SP}}}^4 + \beta_{\mathrm{AF\text{-}SP}}{M_{\mathrm{AF}}}^2{M_{\mathrm{SP}}}^2, \quad (1)$$

where $\alpha_{\mathrm{AF}}(T) = \alpha_{\mathrm{AF}_0}(T - T_{\mathrm{AF}})$ and $\alpha_{\mathrm{SP}}(T) = \alpha_{\mathrm{SP}_0} \times (T - T_{\mathrm{SP}})$. For the sake of simplicity we restrict to the two dominant wave vectors ($\mathbf{Q}$ and $\mathbf{Q}^*$) of the spatial fluctuation only and take their mode-mode coupling into account [14]. We assume that the AF order is dominant ($T_{\mathrm{AF}} > T_{\mathrm{SP}}$) and finally develops long-range order. Once the AF order is established, the SP order is suppressed, since the two magnetic orders compete with each other described by the mode-mode coupling terms ($\beta_{\mathrm{AF\text{-}SP}} > 0$). Thus, only the AF order parameter has a nonzero mean

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value, as the temperature is lowered. Considering the fluctuation effects, we separate the order parameters into mean value and fluctuation part,

$$M_{\mathrm{AF}} = \bar{M}_{\mathrm{AF}} + \delta M_{\mathrm{AF}} \quad \text{and} \quad M_{\mathrm{SP}} = \bar{M}_{\mathrm{SP}} + \delta M_{\mathrm{SP}}, \tag{2}$$

where $\bar{M}_{\mathrm{SP}} = 0$ for all temperatures. The mean value of the AF order parameter is determined by minimizing the free energy with respect to $\bar{M}_{\mathrm{AF}}$, including the renormalization due to fluctuations,

$$\bar{M}_{\mathrm{AF}}^2 = \frac{\alpha_{\mathrm{AF}}(T) + 3\beta_{\mathrm{AF}}\langle \delta M_{\mathrm{AF}}{}^2\rangle + \beta_{\mathrm{AF\text{-}SP}}\langle \delta M_{\mathrm{SP}}{}^2\rangle}{\beta_{\mathrm{AF}}}. \tag{3}$$

The Néel temperature T_N is defined by the vanishing of $\bar{M}_{\mathrm{AF}}^2$. In the calculation of the mean square of the fluctuation in the transition region the renormalization due to the presence of the fourth-order (mode-mode coupling) terms has to be included. This can be done approximately by applying a standard self-consistent decoupling

$$\delta M_{\mathrm{AF}}{}^2 = \langle \delta M_{\mathrm{AF}}{}^2\rangle + (\delta M_{\mathrm{AF}}{}^2 - \langle \delta M_{\mathrm{AF}}{}^2\rangle), \tag{4}$$

$$\delta M_{\mathrm{SP}}{}^2 = \langle \delta M_{\mathrm{SP}}{}^2\rangle + (\delta M_{\mathrm{SP}}{}^2 - \langle \delta M_{\mathrm{SP}}{}^2\rangle), \tag{5}$$

leading to the effective Gaussian fluctuation model [14]. Thus, substituting Eqs. (4) and (5) into the free energy we obtain the following expression up to second order in the fluctuations of the order parameter:

$$\delta\mathcal{F}_M = [\alpha_{\mathrm{AF}}(T) + 3\beta_{\mathrm{AF}}\bar{M}_{\mathrm{AF}}^2 + \beta_{\mathrm{AF}}\langle \delta M_{\mathrm{AF}}{}^2\rangle + \beta_{\mathrm{AF\text{-}SP}}\langle \delta M_{\mathrm{SP}}{}^2\rangle]\delta M_{\mathrm{AF}}{}^2 - \frac{\beta_{\mathrm{AF}}}{2}\langle \delta M_{\mathrm{AF}}{}^2\rangle^2$$
$$+ [\alpha_{\mathrm{SP}}(T) + \beta_{\mathrm{AF\text{-}SP}}\bar{M}_{\mathrm{AF}}^2 + \beta_{\mathrm{SP}}\langle \delta M_{\mathrm{SP}}{}^2\rangle + \beta_{\mathrm{AF\text{-}SP}}\langle \delta M_{\mathrm{AF}}{}^2\rangle]\delta M_{\mathrm{SP}}{}^2 - \frac{\beta_{\mathrm{SP}}}{2}\langle \delta M_{\mathrm{SP}}{}^2\rangle^2. \tag{6}$$

From this we can calculate self-consistently the Gaussian fluctuations including Eq. (3) [14],

$$\langle \delta M_{\mathrm{AF}}{}^2\rangle = \int d(\delta M_{\mathrm{AF}})\delta M_{\mathrm{AF}}{}^2 e^{-\beta\delta\mathcal{F}} \Big/ \int d(\delta M_{\mathrm{AF}}) e^{-\beta\delta\mathcal{F}}, \tag{7}$$

$$\langle \delta M_{\mathrm{SP}}{}^2\rangle = \int d(\delta M_{\mathrm{SP}})\delta M_{\mathrm{SP}}{}^2 e^{-\beta\delta\mathcal{F}} \Big/ \int d(\delta M_{\mathrm{SP}}) e^{-\beta\delta\mathcal{F}}. \tag{8}$$

Since the modulation of the magnetization in the $Ho_{1-x}Dy_xNi_2B_2C$ occurs with a short length scale, i.e., has large wave vector $\mathbf{Q}$ or $\mathbf{Q}^*$, the superconductivity does not respond through coupling to the magnetic field (vector potential). For an (spin singlet) s-wave superconductor the local spin polarization is pair breaking such that the above magnetic order should repel superconductivity for electrons in orbitals subject to a net spin moment due to the $4f$-spin ordering. (A quantitative estimate of this effect requires rather detailed knowledge about the complicated band structure. Interestingly, in experiment the Néel temperature does not deviate much from the de Gennes scaling; i.e., it seems to be basically not affected by the superconducting order. Thus, we neglect the effects of the superconductivity on the magnetism.) Now we can calculate $\langle M_{\mathrm{AF}}^2\rangle$ and $\langle M_{\mathrm{SP}}^2\rangle$ using the above self-consistent scheme. The result of the temperature dependence of the intensities $\langle M^2\rangle$ is shown in Fig. 1. We choose the parameters to obtain a qualitative agreement with the neutron scattering experiments of the pure Ho compound [4]. This yields bare transition temperatures T_{AF} and T_{SP} which differ only by a few percent. Here the SP phase grows as temperature decreases and disappears quickly below the onset of AF order.

According to the band calculation of $LuNi_2B_2C$ [5], the Ni($3d$) band has the largest contribution to the density of states at the Fermi surface suggesting that it dominates in the formation of the superconducting state. However, also the other bands involving hybridizations with B($2p$), C($2p$), and Ho($5d$) or Dy($5d$) should be included in the superconductivity. Hence, superconductivity of the nickelborocarbide materials cannot be described by a single-band model. In connection with the magnetic order there is clear distinction among the bands depending on the location of each element within the unit cell. For the AF order the influence of the rare-earth magnetic moments is canceled on the Ni sites located exactly in the center of a tetrahedron of the nearest Ho (Dy) atoms. The band originating from Ni($3d$) does not feel the magnetic momentum of Ho (Dy) anymore below the Néel temperature. On the other hand, the field generated by the moments of Ho (Dy) is not canceled at the site of the other elements. More-

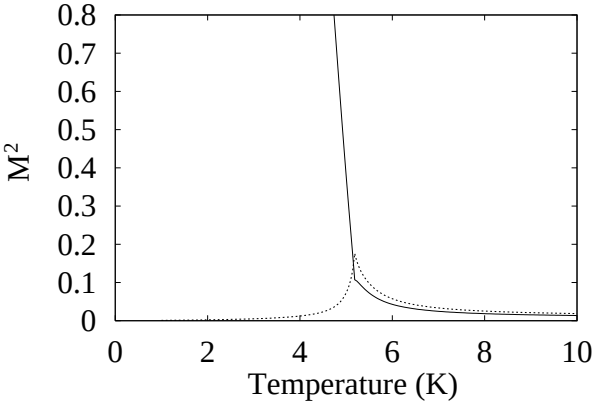

FIG. 1. Intensity of the antiferromagnetic M_{AF} and c-axis spiral M_{SP}. The solid line denotes the intensity of the antiferromagnetic order and the dotted line the intensity of the spiral order.

over, because of large in-plane ferromagnetism of Ho (Dy) magnetic moments, the other ions feel a comparatively strong (pair breaking) spin polarization.

In the case of SP correlation, the magnetic field at the Ni site is not canceled exactly. Thus, the SP phase would also affect the superconductivity of the Ni($3d$) band. For other ions the effect of the SP correlation is not so important since the dominant effect comes eventually from AF order. Hence we propose a simple phenomenological model which consists of two superconducting order parameters. One superconducting order parameter φ_A is associated with the Ni($3d$) band, and couples only to the SP order parameter. The other (φ_B) is due to the other bands involving essentially all the elements. This superconductivity is suppressed by the AF order. The Ginzburg-Landau free energy for these two superconducting order parameters is

$$\mathcal{F}_{\rm SC} = \int d^3r \Big\{ \frac{\hbar^2}{2m_{\rm A}} \Big| \Big(\nabla - \frac{2\pi i}{\phi_0}\vec{A}\Big)\varphi_{\rm A} \Big|^2 + a_{\rm A}(T)|\varphi_{\rm A}|^2 + \frac{1}{2} b_{\rm A}|\varphi_{\rm A}|^4 + \frac{\hbar^2}{2m_{\rm B}} \Big| \Big(\nabla - \frac{2\pi i}{\phi_0}\vec{A}\Big)\varphi_{\rm B} \Big|^2 + a_{\rm B}(T)\,|\varphi_{\rm B}|^2 + \frac{1}{2} b_{\rm B}|\varphi_{\rm B}|^4 - \gamma_1(\varphi_{\rm A}{}^*\varphi_{\rm B} + \text{c.c.}) - \gamma_2\Big[\Big(\nabla + \frac{2\pi i}{\phi_0}\vec{A}\Big)\varphi_{\rm A}{}^*\Big(\nabla - \frac{2\pi i}{\phi_0}\vec{A}\Big)\varphi_{\rm B} + \text{c.c.}\Big] + \eta_1 M_{\rm AF}{}^2|\varphi_{\rm B}|^2 + \eta_2 M_{\rm SP}{}^2|\varphi_{\rm A}|^2 \Big\}, \tag{9}$$

where the parameters are chosen as $T_{cA} = 6.2$ K and $T_{cB} = 2.9$ K (ϕ_0 is standard flux quantum).

For the superconductivity, we obtain two coupled differential equations for superconducting order parameters from the Ginzburg-Landau free energy (9). If we assume Landau gauge, $\mathbf{A} = (0, Hx, 0)$, then the linearization leads to the following homogeneous equations after solving the differential equations:

$$0 = \Big(\frac{\hbar^2}{2m_{\rm A}}\frac{2\pi H}{\phi_0} + a_{\rm A}(T) + \eta_2\langle M_{\rm SP}{}^2\rangle\Big)\varphi_{\rm A} - \Big(\gamma_1 - \gamma_2\frac{2\pi H}{\phi_0}\Big)\varphi_{\rm B}, \tag{10}$$

$$0 = \Big(\frac{\hbar^2}{2m_{\rm B}}\frac{2\pi H}{\phi_0} + a_{\rm B}(T) + \eta_1\langle M_{\rm AF}{}^2\rangle\Big)\varphi_{\rm B} - \Big(\gamma_1 - \gamma_2\frac{2\pi H}{\phi_0}\Big)\varphi_{\rm A}. \tag{11}$$

The superconducting instability corresponds to the vanishing of the determinant of these two equations which determines T_c and H_{c2}. Note that in the absence of an external field the onset of superconductivity occurs at T_c higher than both T_{cA} and T_{cB} as a result of the coupling of the order parameter components.

Now we include the Dy doping in $HoNi_2B_2C$. Dy and Ho have the same magnetic ordering properties and similar magnetic moments except for the magnitude of its dGF which determines the strength of the coupling between localized $4f$-spin and conduction electrons. Thus, we assume that the doping of Dy changes only the average dGF which grows linearly with the Dy concentration x [dGF(x) = dGF(Ho) $\times$ $(1 - x)$ + dGF(Dy) $\times$ x]. We take η_1 and η_2 to be linearly proportional to the dGF. Note that the Néel temperature due to RKKY interaction depends also linearly on the average dGF.

As shown in Fig. 2, our calculation explains well the experimental T_c variation even when $T_c < T_N$. At small doping ($x < 0.2$) the T_c, determined by both the $\varphi_{\rm A}$ and $\varphi_{\rm B}$, decreases due to the AF and SP fluctuations which also introduce an apparent discontinuity of the onset of superconductivity because of a reentrant normal state region near the T_N. For $x > 0.2$ the upper superconducting region ceases to exist. The lower T_c increases slightly with growing T_N and remains as the only superconducting transition for larger Dy-doping concentrations.

Our two component order parameter model gains further support, if we compare the H_{c2} curves of $HoNi_2B_2C$ [15] and $DyNi_2B_2C$ [16]. Figure 3 shows the H_{c2} of our model for $HoNi_2B_2C$ and $DyNi_2B_2C$. Our result reproduces the dip in the upper critical field of the Ho

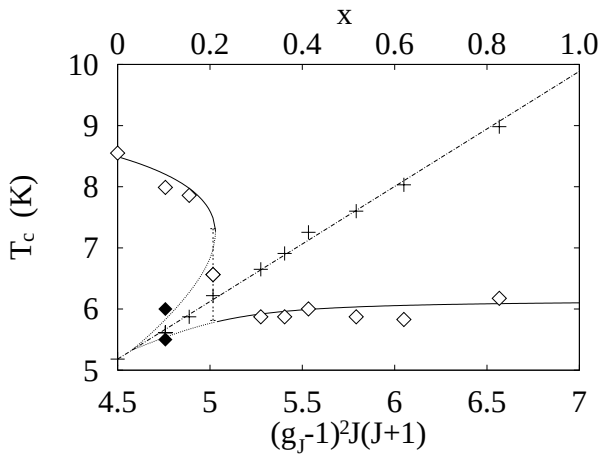

FIG. 2. dGF and doping concentration vs T_c for $Ho_{1-x}Dy_x$-Ni_2B_2C. The dashed line denotes the Néel temperature and the solid line the T_c. The dot-dashed line indicates the phase boundary in the reentrant region. The diamond points mark the T_c, the plus points mark the T_N, and the filled diamond points mark the boundary of the reentrant region in the experiments [10].

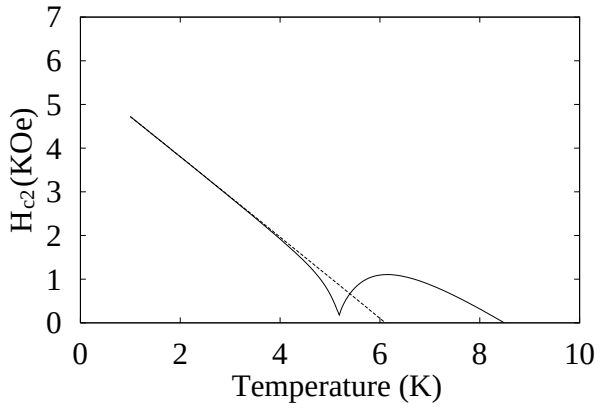

FIG. 3. Temperature vs H_{c2}. The solid line denotes the H_{c2} of $HoNi_2B_2C$, and the dotted line denotes the H_{c2} of $DyNi_2B_2C$.

compound near the Néel temperature as in Ref. [15]. Below the Néel temperature, both H_{c2} curves are more or less identical, because the remaining superconducting (Ni) band feels basically the same magnetic environment in both the Ho and the Dy compounds.

As our model describes some of the physical properties of the alloy $Ho_{1-x}Dy_xNi_2B_2C$ very well, we may ask whether some conclusions can be drawn also for related systems. Starting from $DyNi_2B_2C$, the Dy ions may be replaced by other rare-earth elements. Since the superconductivity in this compound relies on the subtle canceling of the ordered magnetic moments on the Ni site, other elements (different from Ho or Dy) disturb this balance substantially. Thus, doping $DyNi_2B_2C$ with nonmagnetic elements, Lu ($Lu_xDy_{1-x}Ni_2B_2C$), yields a large net moment on the Ni site such that Lu should act like a magnetic impurity. Indeed, the T_c decreases with the doping concentration of Lu in agreement with the Abrikosov-Gor'kov theory. In addition, T_N also drops due to dilution of the Dy moments. A similar behavior is expected if we dope the Dy compound with a magnetic element whose crystal field and spin-orbit coupling effects support a different ordering. Also such dopants disturb the balance and act as a magnetic impurity on the Ni-based superconductivity. Furthermore, these elements introduce disorder in the $4f$-spin system such that simultaneously T_N also decreases with doping concentration. The above alloy gives results consistent with our assumption that the band relevant for superconductivity in the Dy-rich compound is Ni based.

From our model, which describes the behavior and mutual influence of two magnetic and two superconducting order parameters, we can derive a consistent theory explaining all the key experiments in the alloy $Ho_{1-x}Dy_xNi_2B_2C$. The multiband structure is a crucial aspect to understand the superconducting properties in this system [17]. It is an important feature that the Ni($3d$) which dominates the superconductivity does not couple to the spin ordering of the rare earth ions, Ho and Dy. Thus, the mutual interaction of magnetic order and superconductivity is mainly due to the other bands which seem to be weaker superconductors readily suppressed by the magnetism. We have found that our model develops a reentrance behavior near the crossing region of T_c and T_N. This reentrance can also explain qualitatively the nonmonotonic temperature dependence of the electrical resistance for compounds in this doping region. It is basically a consequence of a sequence of a normal-superconducting-normal-superconducting phase as temperature is lowered. Although the reentrant behavior was also reported in polycrystalline $HoNi_2B_2C$, our theory verifies that it appears without any inhomogeneity. Thus the reentrant behavior of $Ho_{1-x}Dy_xNi_2B_2C$ is an intrinsic effect. This region may be very interesting to investigate various fluctuation effects in and close to the superconducting phase. Clearly disorder effects must play an important role here too. Thus experimental studies make sense only with high quality homogeneous samples. In conclusion, we emphasize that, despite the fact that the superconducting phase is probably electron-phonon induced and conventional s-wave type, the physics resulting from the interplay with magnetism and the multiband effects yields a wealth of unusual properties reviving again the study of magnetic superconductors.

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*Present address: Dept. of Material Science and Technology, K-JIST, Kwangju 500-712 Korea.

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