Phenomenological Theory of Superconductivity and Magnetism in Ho\textsubscript{1−x}Dy\textsubscript{x}Ni\textsubscript{2}B\textsubscript{2}C

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The coexistence of the superconductivity and magnetism in the Ho\textsubscript{1−x}Dy\textsubscript{x}Ni\textsubscript{2}B\textsubscript{2}C is studied by using Ginzburg-Landau 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 multi-band 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\textsubscript{1−x}Dy\textsubscript{x}Ni\textsubscript{2}B\textsubscript{2}C showing a reentrant behavior are analyzed yielding a very good agreement with experimental data.

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The discovery of the series of nickelborocarbide, \(RNi_2B_2C\) (\(R = Y, Yb, Dy, Ho, Er, Tm, and Tb\)) \[1\] a few years ago opened a new chapter in the discussion of magnetic superconductors \[2\]. This class of materials shows variety of magnetic ordered states \[3\] 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 \(T_c = 16.6K\) for \(LuNi_2B_2C\). Band calculations of non-magnetic \(LuNi_2B_2C\) \[1\] reveal a complicated multi-band 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 \((g_f - 1)^2J/(J + 1)\) \(dGF\). 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\textsubscript{1−x}Dy\textsubscript{x}Ni\textsubscript{2}B\textsubscript{2}C \[1\]. The experiments show that there is no simple de Gennes scaling of \(T_c\) which is expected to monotonically decrease with increasing \((g_f - 1)^2J(J + 1)\) 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 function of doping concentration \(x\) is apparently discontinuous as a consequence of a reentrant behavior \[1\]. 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\textsubscript{2}B\textsubscript{2}C \[12\] and DyNi\textsubscript{2}B\textsubscript{2}C, show that the magnetic properties of the both materials are highly anisotropic with an easy axis in [110]-direction \[5\] and \[6\]. Both HoNi\textsubscript{2}B\textsubscript{2}C and DyNi\textsubscript{2}B\textsubscript{2}C have antiferromagnetic (AF) order below \(T_N = 5 K\) and 10 K, respectively. The spins order ferromagnetically within the a-b plane and antiferromagnetically in c-direction. For HoNi\textsubscript{2}B\textsubscript{2}C, there exists a significant spiral short-range correlation (SP) with the wave vector, \(Q^c \simeq 0.91Q\), where \(Q\) is the AF wave vector, i.e. the spins are aligned ferromagnetically in the a-b plane, and rotate by \(\phi \sim 163^\circ\) between adjacent planes. A plausible microscopic origin of these two magnetic correlations has been discussed by Kalatsky et al. \[3\] and Amici et al. \[4\].

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,

\[
\mathcal{F}_M = \alpha_{AF}(T)M_{AF}^2 + \alpha_{SP}(T)M_{SP}^2 + \frac{\beta_{AF}}{2}M_{AF}^4 + \frac{\beta_{SP}}{2}M_{SP}^4 + \beta_{AF-SP}M_{AF}^2M_{SP}^2,
\]
where \( \alpha_{\text{AF}}(T) = \alpha_{\text{AF}0}(T - T_{\text{AF}}) \) and \( \alpha_{\text{SP}}(T) = \alpha_{\text{SP}0}(T - T_{\text{SP}}) \). For the sake of simplicity we restrict to the two dominant wave vectors (\( Q \) and \( Q' \)) of the spatial fluctuation only and take their mode-mode coupling into account. We assume that the AF order is dominant \( (T_{\text{AF}} > T_{\text{SP}}) \) 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 term \( (\beta_{\text{AF-SP}} > 0) \). Thus, only the AF order parameter has a non-zero mean value, as the temperature is lowered. Considering the fluctuation effects, we separate the order parameters into mean value and fluctuation part,

\[
M_{\text{AF}} = \bar{M}_{\text{AF}} + \delta M_{\text{AF}} \quad \text{and} \quad M_{\text{SP}} = \bar{M}_{\text{SP}} + \delta M_{\text{SP}} \quad (2)
\]

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

\[
\delta F_M = (\alpha_{\text{AF}}(T) + 3\beta_{\text{AF}}\bar{M}_{\text{AF}}^2 + \beta_{\text{AF}}\delta M_{\text{AF}}^2 + \beta_{\text{AF-SP}}\delta M_{\text{SP}}^2) \delta M_{\text{AF}}^2 - \frac{\beta_{\text{AF}}}{2} \langle \delta M_{\text{AF}}^2 \rangle^2
\]

leading to effective Gaussian fluctuation model \([5]\). Thus, substituting Eq. (3) and (4) into the free energy we obtain the following expression up to second order in the fluctuations of the order parameter,

\[
\bar{M}_{\text{AF}}^2 = \frac{\alpha_{\text{AF}}(T) + 3\beta_{\text{AF}}\delta M_{\text{AF}}^2 + \beta_{\text{AF-SP}}\delta M_{\text{SP}}^2}{\beta_{\text{AF}}} \quad (3)
\]

The Néel temperature \( T_N \) is defined by the vanishing of \( \bar{M}_{\text{AF}}^2 \). 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

\[
\delta M_{\text{AF}}^2 = \langle \delta M_{\text{AF}}^2 \rangle + (\delta M_{\text{AF}}^2 - \langle \delta M_{\text{AF}}^2 \rangle), \quad \delta M_{\text{SP}}^2 = \langle \delta M_{\text{SP}}^2 \rangle + (\delta M_{\text{SP}}^2 - \langle \delta M_{\text{SP}}^2 \rangle),
\]

leading to effective Gaussian fluctuation model \([5]\). Thus, substituting Eq. (3) and (4) into the free energy we obtain the following expression up to second order in the fluctuations of the order parameter.

From this we can calculate self-consistently the Gaussian fluctuations including Eq. (3) \([17]\)

\[
\langle \delta M_{\text{AF}}^2 \rangle = \int d\delta M_{\text{AF}} \delta M_{\text{AF}}^2 e^{-\beta_{\text{AF}}F} / \int d\delta M_{\text{AF}} e^{-\beta_{\text{AF}}F} \quad (7)
\]

\[
\langle \delta M_{\text{SP}}^2 \rangle = \int d\delta M_{\text{SP}} \delta M_{\text{SP}}^2 e^{-\beta_{\text{SP}}F} / \int d\delta M_{\text{SP}} e^{-\beta_{\text{SP}}F} \quad (8)
\]

Since the modulation of the magnetization in the Ho\(_{1-x}\)Dy\(_x\)Ni\(_2\)B\(_2\)C occurs with a short length scale, i.e. has large wave vector \( Q \) or \( Q' \), 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 \( \langle M_{\text{AF}}^2 \rangle \) and \( \langle M_{\text{SP}}^2 \rangle \) 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 \([10]\). This yields a bare transition temperatures \( T_{\text{AF}} \) and \( T_{\text{SP}} \) which differ only by a few percent. Here the SP phase grows as temperature decreases and disappears quickly below the onset of AF order.

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

According to the band calculation of LuNi\(_2\)B\(_2\)C \([11]\), 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 the 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. Moreover, 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 Ni(3d) band.

\[
F_{SC} = \int d^3r \left[ \frac{\hbar^2}{2m_A} |(\nabla - \frac{2\pi i}{\phi_0} \vec{A})\varphi_A|^2 + a_A(T)|\varphi_A|^2 + \frac{1}{2}h_A|\varphi_A|^4 + \frac{\hbar^2}{2m_B} |(\nabla - \frac{2\pi i}{\phi_0} \vec{A})\varphi_B|^2 + a_B(T)|\varphi_B|^2 + \frac{1}{2}h_B|\varphi_B|^4 \right] - \gamma_1(\varphi_A^\dagger \varphi_B + c.c) - \gamma_2 \left[ (\nabla + \frac{2\pi i}{\phi_0} \vec{A})\varphi_A^\dagger(\nabla - \frac{2\pi i}{\phi_0} \vec{A})\varphi_B + c.c \right] + \eta_1 M_{AF}^2 |\varphi_B|^2 + \eta_2 M_{SP}^2 |\varphi_A|^2, \tag{9}
\]

where the parameters are chosen as \(T_{cA} = 6.2\,\text{K}\), and \(T_{cB} = 2.9\,\text{K}\) (\(\phi_0\) : standard flux quantum).

For the superconductivity, we obtain two coupled differential equations for superconducting order parameters

\[
0 = \left( \frac{\hbar^2}{2m_A} \frac{2\pi H}{\phi_0} + a_A(T) + \eta_2 (M_{SP}^2) \right) \varphi_A - \left( \gamma_1 - \gamma_2 \frac{2\pi H}{\phi_0} \right) \varphi_B \tag{10}
\]

\[
0 = \left( \frac{\hbar^2}{2m_B} \frac{2\pi H}{\phi_0} + a_B(T) + \eta_1 (M_{AF}^2) \right) \varphi_B - \left( \gamma_1 - \gamma_2 \frac{2\pi H}{\phi_0} \right) \varphi_A \tag{11}
\]

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\(_2\)B\(_2\)C. Dy and Ho has 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 the doping of Dy changes only the average dGF which grows linear with the Dy concentration \(x\) (dGF\((x) = \text{dGF(Ho)} \times (1-x) + \text{dGF(Dy)} \times x\)). We take \(\eta_1\) and \(\eta_2\) 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 \(\varphi_A\) and \(\varphi_B\), 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.

**FIG. 2.** dGF and doping concentration vs. \(T_c\) for Ho\(_{1-x}\)Dy\(_x\)Ni\(_2\)B\(_2\)C. 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 the \(T_N\), and the filled diamond points the boundary of the reentrant region in the experiments [11].

Our two component order parameter model gains fur-
ther support, if we compare the $H_{c2}$ curves of HoNi$_2$B$_2$C [14] and DyNi$_2$B$_2$C [17]. Fig.3 shows the $H_{c2}$ of our model for HoNi$_2$B$_2$C and DyNi$_2$B$_2$C. Our result reproduces the dip in the upper critical field of Ho-compound near the Néel temperature as in Ref. [14]. 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 Dy-compound.

![Graph showing $H_{c2}$ vs. temperature for HoNi$_2$B$_2$C and DyNi$_2$B$_2$C.](image)

FIG. 3. Temperature vs. $H_{c2}$. The solid line denotes the $H_{c2}$ of HoNi$_2$B$_2$C, and the dotted line denotes the $H_{c2}$ of DyNi$_2$B$_2$C.

As our model describes some of the physical properties of the alloy Ho$_{1-x}$ Dy$_x$Ni$_2$B$_2$C very well, we may ask whether some conclusions can be drawn also for related systems. Starting from DyNi$_2$B$_2$C, 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$_2$B$_2$C with non-magnetic elements, Lu, (Lu$_x$Dy$_{1-x}$Ni$_2$B$_2$C) 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$_x$Ni$_2$B$_2$C. The multi-band structure is a crucial aspect to understand the superconducting properties in this system. 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 non-monotonic temperature dependence of the electrical resistance for compounds in this doping region. It is basically a consequence of a sequence of normal-superconducting-normal-superconducting and vice versa. The model was also applied to polycrystalline HoNi$_2$B$_2$C, where it was verified that it appears without any inhomogeneity. Thus the reentrant behavior of Ho$_{1-x}$ Dy$_x$Ni$_2$B$_2$C 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 only sense with high quality homogeneous samples. In conclusion, we would like to 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 multi-band effects yield a wealth of unusual properties reviving again the study of magnetic superconductors.

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