Competing magnetic and superconducting order in the rare-earth borocarbides RNi$_2$B$_2$C (R=Tm, Er, Ho, Dy)

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The competition between superconductivity and magnetic ordering in the rare-earth borocarbides is analyzed in terms of an estimate of the different contributions to the free energy. The two most important effects are the Anderson-Suhl screening [Phys. Rev. **116**, 898 (1959)] of the indirect exchange interaction and the reduction of the number of Cooper-pair states by the superzone gaps created by the antiferromagnetic ordering. In the case of TmNi$_2$B$_2$C, the theory accounts for the anisotropy of the upper critical field, the field dependence of the radius of the vortices, and the jump in the derivative of the bulk magnetization at the superconducting transition. The theory also gives a fair account of the magnetic effects on the upper critical fields in the Er, Ho, and Dy borocarbides.

I. INTRODUCTION

The rare-earth borocarbides RNi$_2$B$_2$C, with R=Tm, Er, Ho, and Dy, show the exceptional property of superconductivity coexisting and competing with antiferromagnetic ordering, and they may be classified as unconventional superconductors in their own right. The crystal structure is tetragonal with the rare-earth ions placed on a body-centered lattice. Recent and comprehensive descriptions of these systems may be found in Refs. [1–3]. The two electronically equivalent, but nonmagnetic, compounds LuNi$_2$B$_2$C and YNi$_2$B$_2$C are both strong type-II superconductors below T_c ≃16 K, and the upper critical field in the zero-temperature limit $H_{c2}(0)$ is 80–110 kOe, when the field is applied parallel to the c axis. [4,5] It is normally assumed that the most important reason for the suppression of the superconducting phase in a homogeneous magnetic system is the existence of low-energy thermal fluctuations of the magnetic moments. That this cannot be the whole story is clear from the observation that whereas the transition temperature in the Tm compound is only reduced by about 30% in comparison with the two nonmagnetic systems, the upper critical field $H_{c2}(0)$ along the c axis is a factor of 10 smaller. Additionally, the anisotropy of H_{c2} is much larger than the 15% observed in the Lu and Y systems. Here, we shall concentrate on the two alternative mechanisms presented below for explaining these and similar features observed in the other three rare-earth systems.

The localized magnetic rare-earth moments are coupled indirectly through the conduction electrons. The ferromagnetic component of this Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange interaction between the rare-earth moments is reduced in proportion to the superconducting order, because of the Anderson-Suhl screening of the electron-spin susceptibility in the long wavelength limit. [6] The screening is not due to currents, as in the case of the magnetic field, but occurs because the Bardeen-Cooper-Schrieffer (BCS) ground state is a singlet. The screening implies that the upper critical field is going to depend on the magnetization induced by the applied field and thereby on the anisotropy of the magnetic system. As first discussed in this connection in Ref. [7], the magnitude of the screening, in the case of the Tm compound, is determined by the derived stability of the antiferromagnetic ordering in the presence of a c-axis field. Due to low-lying electronic excitations associated with the vortices, the screening is expected to be absent within the cores of the vortices in the type-II phase. This implies an excess magnetization of the flux lines in comparison with their surroundings and a radius of the cores that increases with field.

Because of the RKKY exchange, the properties of the Fermi surface are affected by a magnetic ordering, and any change of the Fermi surface may change the superconducting state. [8,9] A sinusoidally magnetic ordering gives rise to a new periodicity of the system, and the RKKY interaction between the spins of the 4f electrons and the conduction electrons leads to energy gaps at the wave vectors ($\boldsymbol{\tau}\pm n\boldsymbol{Q}$)/2, where $\boldsymbol{\tau}$ is a reciprocal lattice vector and $\boldsymbol{Q}$ is the magnetic ordering wave vector. If a superzone energy gap cuts through the Fermi surface, the lifting of the spin degeneracy implies that the states near the gap become unavailable for the Cooper pairs, leading to a reduction of the "effective density of states" at the Fermi surface and thereby to a reduction of the superconducting energy gap.

In the next section, we present a free-energy analysis of the competition between superconducting and magnetic ordering. The theory is then applied in succession to the four rare-earth borocarbides in Sec. III with the main focus on TmNi$_2$B$_2$C. The conclusions are given in Sec. IV.

II. THEORETICAL MODEL

With the basic assumption that the borocarbides are standard BCS systems, the theory [6] determines the zero-frequency susceptibility at zero temperature to be

$$\chi_s^0(\boldsymbol{q}) = \frac{1}{8N}\sum_{\boldsymbol{k}} \frac{(\xi_{\boldsymbol{k}-\boldsymbol{q}} - \xi_{\boldsymbol{k}})^2 - (E_{\boldsymbol{k}-\boldsymbol{q}} - E_{\boldsymbol{k}})^2}{E_{\boldsymbol{k}-\boldsymbol{q}}E_{\boldsymbol{k}}(E_{\boldsymbol{k}-\boldsymbol{q}} + E_{\boldsymbol{k}})}, \quad (1)$$

where $\xi_{\boldsymbol{k}}=\varepsilon_{\boldsymbol{k}}-\mu$ is the energy of the conduction electrons at wave vector $\boldsymbol{k}$ subtracted the chemical potential, and $E_{\boldsymbol{k}} = \sqrt{\xi_{\boldsymbol{k}}^2+\Delta^2}$. The superconducting gap Δ is determined by
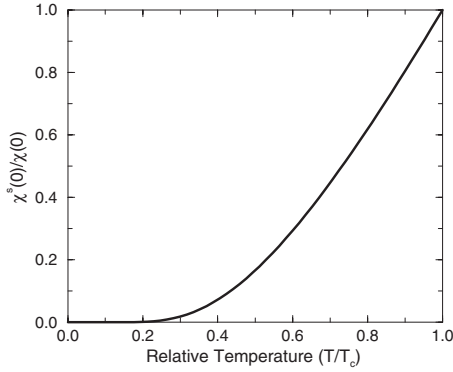
FIG. 1. The temperature dependence of the electronic susceptibility in the superconducting phase relative to the normal-phase value.

$$\frac{1}{\mathcal{N}(0)g_s} = \ln\left(\frac{2\hbar\omega_D}{\Delta(0)}\right) = \int_0^{\hbar\omega_D} \frac{\tanh\frac{1}{2}\beta(x^2+\Delta^2)^{1/2}}{(x^2+\Delta^2)^{1/2}}dx, \quad (2)$$

where $\mathcal{N}(0)$ is the density of states per unit cell and per spin state at the Fermi surface, g_s is the electron-phonon coupling parameter, ω_D is the Debye frequency, and $\Delta(0)$ $=1.764\ k_BT_c$. The temperature dependence of the zero-frequency susceptibility at zero wave vector, relative to the susceptibility $\chi(\mathbf{0})=\mathcal{N}(0)/2$ of the normal phase, is then determined by the integral (assuming $T \ll T_F$)

$$\chi_s(\mathbf{0})/\chi(\mathbf{0}) = 2\int_0^\infty \frac{e^{\sqrt{x^2+(\beta\Delta)^2}}}{\left[e^{\sqrt{x^2+(\beta\Delta)^2}}+1\right]^2}dx. \quad (3)$$

The integral vanishes exponentially, $\propto\exp[-\beta\Delta(0)]$, at low temperatures and approaches 1 linearly when $T\rightarrow T_c$. A numerical evaluation of this integral is shown in Fig. 1. The field $h=2\mu_B(H+H_{\mathrm{ex}})$ acting on the electronic spin components has to be a substantial fraction of $\Delta(0)$ before it modifies the zero-field results noticeably. The part due to the applied field in the type-II phase is of no importance. The exchange field due to the polarization of the $4f$ moments of the rare-earth ions may be of some significance; however, since the effects on the susceptibility are of second order in h, the field dependence of $\chi_s(\mathbf{0})$ is neglected.

Linearizing the energies near the Fermi surface, $\xi_{\mathbf{k}}$ $=\hbar v_F(|\mathbf{k}|-k_F)$, the integral in Eq. (1) may be calculated numerically. At the smallest values of q, the susceptibility increases as q^2, but in a large interval (as long as $0 < q \ll k_F$), the result is

$$\chi_s^0(\mathbf{q})/\chi(\mathbf{0}) = 0.99\frac{q}{q+1.5q_0}, \quad q_0 = \frac{\pi\Delta}{\hbar v_F} \approx \frac{1}{\xi}, \quad (4)$$

where ξ is the superconducting correlation length. Hence, the zero-temperature susceptibility of the superconductor starts out at zero but is closely approaching the normal-phase value already when $q \simeq 10\xi^{-1} \approx 0.1$ Å^{-1}.

The simplest free-electron model leads to an RKKY interaction[10]

$$\mathcal{H}_{\mathrm{H}} = \frac{1}{2}\sum_{ij}\mathcal{J}(ij)\mathbf{J}_i\cdot\mathbf{J}_j, \quad \mathcal{J}(\mathbf{q})=\sum_j\mathcal{J}(ij)e^{-i\mathbf{q}\cdot\mathbf{R}_{ij}}, \quad (5)$$

with $\mathcal{J}(\mathbf{q})\propto(g-1)^2[\chi(\mathbf{q})-\Sigma_k\chi(\mathbf{k})]$, where g is the Landé factor of the rare-earth ions. Because the electronic susceptibility is modified in the superconducting phase, the RKKY interaction is also changed. Based on the analysis above, we assume that the RKKY interaction in the superconducting phase is

$$\mathcal{J}_s(\mathbf{q}) = \mathcal{J}(\mathbf{q}) - \Delta\mathcal{J}[1-\chi_s(\mathbf{0})/\chi(\mathbf{0})]\delta_{\mathbf{q0}}, \quad (6)$$

i.e., that $\mathcal{J}_s(\mathbf{q})$ is unchanged unless $\mathbf{q}\equiv\mathbf{0}$. $\mathcal{J}(\mathbf{0})$ is only partly determined by $\chi(\mathbf{0})$, and even the simplest model above predicts $\Delta\mathcal{J}$ to be different from $\mathcal{J}(\mathbf{0})$. For instance, $\Delta\mathcal{J}$ has to be positive, which is not necessary the case for $\mathcal{J}(\mathbf{0})$. In the analysis below, $\Delta\mathcal{J}$ is considered to be an adjustable positive parameter, and $\chi_s(\mathbf{0})/\chi(\mathbf{0})$ is assumed to be determined by Eq. (3), i.e., to depend on T but to be independent of an applied field.

The modification of the RKKY interaction given by Eq. (6) is only significant when the spatial average of $\langle J_z \rangle$ is nonzero as induced by an applied field along the z direction. The free energy of the superconducting phase is then increased by (approximately) $\frac{1}{2}[\mathcal{J}(\mathbf{0})-\mathcal{J}_s(\mathbf{0})]\langle\langle J_z\rangle\rangle^2$ per rare-earth ion in comparison with the normal phase. This loss in magnetic energy of the superconducting phase implies a reduction of the upper critical field in comparison with its nonmagnetic value. By being proportional to $\langle\langle J_z\rangle\rangle^2$, the reduction is going to depend sensitively on how large the induced uniform moment is at a certain field, i.e., whether the field is along a magnetically hard or easy axis. Macroscopically, the Anderson-Suhl screening reduces the upper critical field because of its effect on the energy balance between the two phases. On the microscopic scale of a type-II system, the "normal" and the superconducting phases are mixed, in the sense that the magnetic screening disappears within the cores of the vortices due to the presence of low-lying spin-polarizable excitations. Assuming the cores to be normal with respect to the magnetic interactions, the radius of the vortices $\propto\xi$ is going to be larger than in the nonmagnetic case, because of the extra magnetic energy gain of the cores, and the upper critical field $H_{c2}=\Phi_0/(2\pi\xi^2)$ is consequently reduced (Φ_0 is the magnetic flux quantum of the Cooper pairs). The allowance of the superconducting and the magnetically normal phases to mix in this way implies that the transition at the upper critical field becomes of second order in accordance with experiments (see, for instance, Ref. 11). In the previous analysis reported in Ref. 7, the two phases were for simplicity considered to be completely separated, in which case the transition is a first-order one.

In order to quantify the reduction of the upper critical field due to the phase dependence of the RKKY exchange, we need to compare the different contributions to the free energy. The properties of the nonmagnetic version of the type-II superconductor are assumed to be specified in terms of a simplified version of the Ginzburg-Landau free-energy expression

$$F = a\langle|\psi|^2\rangle + \frac{1}{2}b\langle|\psi|^2\rangle^2 + F^n \qquad (7)$$

per formula unit, where the kinetic energy is included in an indirect fashion in terms of the effective squared order parameter $\langle|\psi|^2\rangle$. The requirements to be met by this free energy are that the second-order transition to the normal phase should occur at H_{c2} and that the magnetic susceptibility of the nonmagnetic type-II superconductor should be the negative of

$$X_\kappa = \{1.16[4\pi(2\kappa^2 - 1)N]\}^{-1}, \qquad (8)$$

as derived from the free energy of the hexagonal Abrikosov vortex lattice,[12] where $\kappa = \lambda/\xi$ is the Ginzburg-Landau parameter. In the simplest approximation, κ and b may be considered to be independent of field and temperature, in which case the two requirements determine the two energy parameters to be

$$a = X_\kappa(H_i - H_{c2})H_{c2}(0), \quad b = X_\kappa[H_{c2}(0)]^2, \qquad (9)$$

where $H_{c2} = H_{c2}(T)$ is the upper critical field at the temperature T of the nonmagnetic system and $H_{c2}(0)$ is the zero-temperature value. H_i is the internal field acting on the conduction electrons. In the nonmagnetic case, H_i is equal to the applied field to a good approximation, at least at fields larger than H_{c2}/κ, where the demagnetization field of the diamagnetic moment is negligible.

This phenomenological theory gives a good estimate of the free energy of the superconducting phase. This is obviously true when the applied field is close to the upper critical field, but it is also valid in the limit of zero field, where the result is $F - F^n = -\frac{1}{2}X_\kappa H_{c2}^2 \approx -H_c^2/(8\pi)$, since the critical field $H_c = H_{c2}/(\sqrt{2}\kappa)$ and $\kappa^2 \gg 1$ in the present systems (κ is lying in the range of 5–12). The zero-field value of the squared order parameter is predicted to be

$$|\psi_0|^2 \equiv \langle|\psi|^2\rangle|_{H=0} = \frac{H_{c2}}{H_{c2}(0)}, \qquad (10)$$

which appears to be an acceptable estimate of the temperature dependence of the relative squared order parameter of the homogeneous superconductor at zero field. Hence, the parametrization of the free energy given by Eqs. (7)–(9) gives a reasonable overall description of a nonmagnetic superconductor in the limit of $\kappa^2 \gg 1$.

When the magnetic ions of the superconducting system are introduced, the free energy is assumed to be

$$F = a\langle|\psi|^2\rangle + \frac{1}{2}b\langle|\psi|^2\rangle^2 + (F_M^s - F_M^n)\frac{\langle|\psi|^2\rangle}{|\psi_0|^2} + F_M^n. \quad (11)$$

Here, F_M^s is the free energy per rare-earth ion as derived from the magnetic Hamiltonian of the rare-earth ions in the case of a homogeneous superconductor at the temperature T and field H, and F_M^n is the corresponding free energy of the normal phase. The magnetic Hamiltonian in the superconducting case only differs from the normal one by $\mathcal{J}(\mathbf{0})$ being replaced by $\mathcal{J}_s(\mathbf{0})$. At zero field F_M^n cancels out, and F_M^s is independent of $|\psi_0|^2$ except for the remarkable case of Er borocarbide below about 2.3 K, where there is a small ferromagnetic

component.[13,14] However, the additional $|\psi_0|^2$ dependence introduced by the $\Delta\mathcal{J}$ energy term in this exceptional case is negligible and Eq. (10) still applies. The magnetization of the sample induced by the applied magnetic field H contributes to the effective internal field acting on the superconducting electrons, i.e., H_i in Eq. (9) is now

$$H_i = H + H_D^0(1 - D_z)\langle\langle J_z\rangle\rangle, \quad H_D^0 = 4\pi g\mu_B N, \qquad (12)$$

where D_z is the (relative) demagnetization factor. Neglecting the small diamagnetic contribution of the superconducting electrons, the free-energy expression (11) predicts the average magnetization to be

$$\langle\langle J_z\rangle\rangle = \frac{\langle|\psi|^2\rangle}{|\psi_0|^2}\langle\langle J_z\rangle\rangle_s + \left(1 - \frac{\langle|\psi|^2\rangle}{|\psi_0|^2}\right)\langle\langle J_z\rangle\rangle_n, \qquad (13)$$

where $\langle\langle J_z\rangle\rangle_{s(n)}$ is the site average of $\langle J_z\rangle$ in the pure superconducting (normal) phase. Like Eq. (7) in the nonmagnetic case, the free-energy expression (11) predicts the transition between the type-II and the normal phases to be of second order, and the applied upper critical field is derived to be

$$H = H_{c2}^\star - H_D^0(1 - D_z)\langle\langle J_z\rangle\rangle_n,$$

$$H_{c2}^\star = H_{c2} - \frac{F_M^s - F_M^n}{X_\kappa H_{c2}}. \qquad (14)$$

The result (13) for the average magnetization indicates that the ratio $\langle|\psi|^2\rangle/|\psi_0|^2$ may roughly be interpreted as the volume of the pure superconducting phase, with the order parameter $|\psi_0|$, relatively to the total volume of the mixed type-II phase, and, correspondingly, $1 - \langle|\psi|^2\rangle/|\psi_0|^2$ as the relative volume of the normal-phase cores of the vortices. A purely phenomenological approach suggests that $\langle|\psi|^2\rangle/|\psi_0|^2$ in Eq. (11) may be multiplied by $f(\langle|\psi|^2\rangle)/f(|\psi_0|^2)$, where $f(x)$ is an arbitrary function. In terms of a Taylor expansion, the leading order correction is obtained from $f(x) = 1 + \nu x$, in which case the last term in the expression for $H_{c2}^\star$ is being multiplied by $[1 + \nu H_{c2}/H_{c2}(0)]^{-1}$. Hence, the result for the critical field in Eq. (14) should be trustworthy close to T_c but may be questioned at the lowest temperatures. That the result is correct close to T_c can be verified by a direct standard calculation[12] of $H_{c2}^\star$ utilizing that $1 - \chi_s(\mathbf{0})/\chi(\mathbf{0}) = 2(1-t)/t^2$ in this limit ($t = T/T_c$).

The second possibility for the RKKY interaction to affect the superconducting state derives from the superzone energy gaps produced by the antiferromagnetic order parameter. This effect has been discussed several times in the literature (see, for instance, Nass et al.[8]) and most recently by Amici et al.,[9] who considered the phase diagram of $HoNi_2B_2C$. The leading order effect is that the density of states $\mathcal{N}(0)$ in the gap equation (2) is being reduced proportional to the superzone energy gaps. The largest of these derives from the first harmonic of the antiferromagnetic ordering. The superzone gaps are proportional to the amplitude $\langle J_z(\mathbf{Q})\rangle$ of this harmonic, and we simply assume that $\mathcal{N}(0)$ in Eq. (2) is being replace by

$$\mathcal{N}_{\text{eff}}(0) = \mathcal{N}(0)(1 - \delta), \quad \delta = d_Q \langle J_z(\boldsymbol{Q}) \rangle. \quad (15)$$

The adjustable parameter d_Q introduced here is proportional to the RKKY exchange coupling parameter and is going to depend strongly on the wave vector of the first harmonic. The modification of the gap equation implies that the transition temperature T_c is reduced in comparison with the nonmagnetic value T_c^0 according to

$$\frac{T_c}{T_c^0} = \frac{\Delta(0)}{\Delta_0(0)} = \exp\left[\frac{\delta}{1 - \delta} \ln\left(\frac{\Delta_0(0)}{\hbar \omega_D} \right) \right] \equiv \gamma. \quad (16)$$

The relation between δ and γ is assumed to be $\gamma = \exp[3.6\delta/(1 - \delta)]$ in all the four borocarbides. The precise value of the constant is not important, but the logarithmic term is found to be 3.6 when using $\Delta_0(0) = 1.7$ meV and $\hbar \omega_D = 32$ meV. The latter value corresponds to a Debye temperature of 370 K, as indicated by heat capacity results.

The scaling introduced by Eq. (16) implies a reduction of the energy gained by the superconducting ordering at zero temperature by a factor γ^2, or that $H_c(0) \propto \gamma$. Considering that this scaling has only a minor or no effect on the London penetration depth $\lambda(0)$ but changes $\xi(0)$ by the factor γ^{-1}, we arrive at

$$T_c = \gamma T_c^0, \quad \kappa = \gamma \kappa_0, \quad H_{c2}(0) = \gamma^2 H_{c2}^0(0), \quad (17)$$

where $H_{c2}^0(0)$ and κ_0 are the nonmagnetic values of these parameters. Notice that $\langle J_z(\boldsymbol{Q}) \rangle$, and thereby γ, depends on temperature and fields, implying that T_c, X_κ, and $H_{c2}(0)$ are now functions of the thermodynamic variables in the antiferromagnetic phase.

III. UPPER CRITICAL FIELD

The theory developed in Sec. II is applied on the four superconducting rare-earth borocarbides in the following sections. The theory is parametrized in terms of the "nonmagnetic" constants, T_c^0 and κ_0, and the temperature dependent upper critical field. These parameters are, in principle, those determined in the nonmagnetic counterparts Lu and Y borocarbides,[4,5] i.e., $T_c^0 \simeq 16$ K and $\kappa_0 \approx 12$. The critical field when the field is applied perpendicular to the c axis is assumed to be

$$H_{c2} = \gamma^2 H_{c2}^0(0)[1 - 0.875t - 0.892t^2 + 0.767t^3], \quad (18)$$

where $t = T/T_c = T/(\gamma T_c^0)$ and $H_{c2}^0(0) \simeq 130$ kOe. There are some differences between the experimental upper critical fields of the Lu and Y compounds, especially in the zero-temperature limit, and the variation assumed in Eq. (18) is an average of the experimental results obtained in the two systems. The experimental results also show an anisotropy between the cases where the field is applied either perpendicular or parallel to the c axis. Accordingly, we assume in all four cases discussed below that $H_{c2}^0(0)$ is reduced by 15% when the field is applied parallel to the c axis. Although the electronic properties of the different rare-earth borocarbides presumably are very similar, there are certainly differences of some importance, and we have adjusted T_c^0 and $H_{c2}^0(0)$ to the experimental situation of each case. In order to be consistent with the scaling in Eq. (17), κ_0^2 is scaled with the same factor as applied in the case of $H_{c2}^0(0)$. The two remaining parameters of the theory are $\Delta \mathcal{J}$ in Eq. (6) and d_Q in Eq. (15). The two coupling constants should, in principle, scale with, respectively, $(g-1)^2$ and $(g-1)$, but have been adjusted freely for each system.

The internal field H_i in Eq. (12), and the corresponding field $H - D_z H_D^0 \langle\langle J_z \rangle\rangle$ acting on the magnetic ions, is assumed to be uniform within the type-II phase. In principle, the B field inside the flux lines is shielded from the surroundings, and should be stronger in the magnetically normal phase of the cores compared to the field in the superconducting volume between the vortices. However, because of the large value of κ, the London penetration depth λ is large ($\gtrsim 700$ Å) and the variation of B_i is less than 10% already at a field of ~ 2 kOe. Certainly, the spatial variation has completely vanished when approaching the upper critical field, and the assumption of a uniform internal field is fulfilled in most of the applications below. Since the internal fields include significant contributions from the magnetization of the samples, it is important to have a reasonable estimate of the demagnetization factor. Most of the samples used in the different experiments are flat disks oriented perpendicular to the c axis. Hence, in most cases $D_c \approx 0.7$ and $D_a \approx D_b \approx (D_c - 1)/2$.

The magnetic Hamiltonian of the rare-earth ions is known with reasonable accuracy for all four borocarbides. Gasser *et al.*[15,16] have determined the crystal-field Hamiltonian for each of the different compounds by combining the results for the crystal-field level intensities, observed in inelastic neutron scattering experiments, with susceptibility measurements. A thorough discussion of the different antiferromagnetic structures observed by neutron diffraction in the different rare-earth borocarbides is presented by Lynn *et al.*[17]

A. TmNi$_2$B$_2$C

The magnetic properties of $TmNi_2B_2C$ have been studied by neutron diffraction in detail.[7,18–21] The system orders antiferromagnetically at $T_N = 1.52$ K. The ordering wave vector is $\boldsymbol{Q}_F = (0.094, 0.094, 0)$ (in reciprocal lattice units) with the moments polarized along the c axis. As discussed in Ref. 7, the small value of $|\boldsymbol{Q}_F|$ is readily explained as being an effect of the Anderson-Suhl screening. Furthermore, the antiferromagnetic ordering, at 0.1 K, stays stable as long as the system is superconducting up to a c-axis field of about $H_N \simeq 8$ kOe (except that the ordering wave vector changes its direction above 2 kOe).[20,21] The value of T_N implies that $\mathcal{J}(\boldsymbol{Q}_F) \simeq 8.6$ μeV. The stability field $H_N - D_z H_D^0 \langle\langle J_z \rangle\rangle$ times $g\mu_B$ is a measure of the energy difference $[\mathcal{J}(\boldsymbol{Q}_F) - \mathcal{J}_s(\boldsymbol{0})] \times \langle J_z \rangle$. Since, in the normal phase $\mathcal{J}(\boldsymbol{Q}_F) \approx \mathcal{J}(\boldsymbol{0})$ because of the small value of $|\boldsymbol{Q}_F|$, this estimate leads to $\Delta \mathcal{J}$ lying between 5 and 8 μeV or $\Delta \mathcal{J} \approx 6.5$ μeV, as used in the model calculations.

At the lowest temperatures, the $\boldsymbol{Q}_F$ ordering disappears, when applying a field of 10–15 kOe in the a direction, and is replaced by a new antiferromagnetic ordering of the c-axis moments at the wave vector $\boldsymbol{Q}_A = (0.483, 0, 0)$.[7,18] Against any intuitive anticipations, this antiferromagnetic ordering
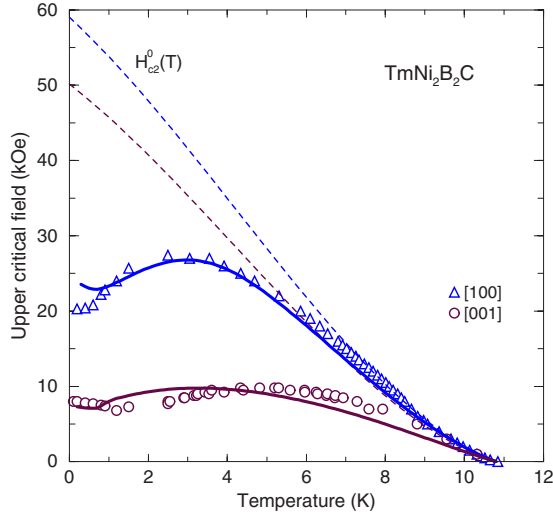
FIG. 2. (Color online) Upper critical field in TmNi$_2$B$_2$C. The experimental results are from Ref. 22. The dashed lines show the assumed values of $H_{c2}^0(T)$ [the temperature factor is the one of H_{c2} in Eq. (18) with $\gamma = 1$], when the field is parallel or perpendicular to c. The solid lines are the calculated results using $D_c = 0.64$.

TABLE I. The model parameters of the four rare-earth borocarbides. The nonmagnetic upper critical field $H_{c2}^0(0)$ is the value used, when the applied field is perpendicular to the c axis. $H_{c2}^0(0)$ is changed by the factor of 0.85 of in the c-axis case.

	Tm	Er	Ho	Dy
$g-1$	1/6	1/5	1/4	1/3
T_c^0 (K)	10.8	11.2	8.8	16
$H_{c2}^0(0)$ (kOe)	59	58	30	74
κ_0	6.9	6.8	4.9	
$\Delta\mathcal{J}$ (μeV)	6.5	3.6	2.0	
d_Q	0.018	0.007	0.013	0.029

becomes more stable the larger the field. This surprising observation has now been explained by the discovery that the system is in a quadrupolar ordered phase below $T_Q = 13.5$ K, at zero field, showing up as a modulated c-axis displacement of the Tm ions at the wave vector $\boldsymbol{Q}_A$.[19] The antiferromagnetic ordering is induced by the uniform magnetic field due to the oscillating anisotropy term associated with the quadrupolar ordering. An important property of the quadrupolar phase is that the order parameter is extremely sensitive to the a-axis field, and at low temperatures it is enhanced by a factor of 10 in a field of 100 kOe compared to the zero-field value. When the field is applied along the c axis, the quadrupolar order parameter stays small and only induces a minute, longitudinally polarized, antiferromagnetic moment. The quadrupolar ordering does not have much influence on the upper critical field in the c-axis case, not only because the order parameter stays small but also because the superzone energy gaps produce by the quadrupolar moments do not affect the spin degeneracy of the conduction electrons, in which case the changes of $\mathcal{N}(0)$ and of the effective $\mathcal{N}(0)$ in the gap equation are the same, and they are only of second order in the energy gaps. When the field is applied perpendicular to the c axis, the antiferromagnetic ordering induced by the field enhances the superzone energy gaps and gives rise to a linear reduction of the effective $\mathcal{N}(0)$. In practice, the effects of the two contributions to the superzone gaps may hardly be distinguished, and the extra superzone effects due to the quadrupolar ordering are considered to be accounted for by introducing an effective coupling parameter d_Q multiplying the amplitude of the modulated antiferromagnetic moments in Eq. (15).

The calculated upper critical fields in the case of TmNi$_2$B$_2$C are compared with experiments in Fig. 2. The results are obtained using the parameters given in Table I and the magnetic Hamiltonian developed in Ref. 19. In the a-axis case, the amplitude of the $\boldsymbol{Q}_A$ antiferromagnetic ordering

grows smoothly as the temperature is reduced and is only of importance for the upper critical field below about 5 K. In the case where the field is along the c axis, the superzone effects due to the $\boldsymbol{Q}_F$ ordering, below ~ 1 K, are small because the amplitude $\langle J(\boldsymbol{Q}_F) \rangle$ is nearly zero close to the upper critical field. In all circumstances, they are assumed to be dominated by the small extra energy gained in the superconducting phase, because it stabilizes the antiferromagnetic $\boldsymbol{Q}_F$ ordering. This implies a small increase of the upper critical field below 1 K, in agreement with observations.

At the transition from the superconducting to the normal phase, the theory predicts a continuous variation of the magnetization, but the derivative of the magnetization is changed by

$$\left. \frac{\Delta M}{\Delta H} \right|_{s+n} - \left. \frac{\Delta M}{\Delta H} \right|_n = \frac{g\mu_B(\langle\langle J_z\rangle\rangle_s - \langle\langle J_z\rangle\rangle_n)}{H_{c2}} \left(\frac{\partial H_{c2}^*}{\partial H} - 1 \right)$$

$$= \frac{g\mu_B(2.845 - 3.518)}{40.7\text{kOe}}(-4.29)$$

$$= 0.08\mu_B/\text{kOe} \qquad (19)$$

at 2 K, when the field is along the c axis. This value agrees acceptably with the experimental observation by Cho $et\ al.$[11] of a jump of about 0.05 μ_B/kOe. In the case that the field is applied perpendicular to the c axis, both the calculated and the experimental values are a factor of about 7 smaller.

Eskildsen $et\ al.$[23] have measured the neutron cross sections of the vortices in Y and Lu borocarbides. The form factor of the (1,0) reflection of the flux-line lattice is determined by

$$|h_{10}|^2 = \frac{\Phi_0^2}{(2\pi\lambda)^4} e^{-4\pi^2 B_i \xi^2/\Phi_0}, \qquad (20)$$

from which they derived $\xi \simeq 85$ Å and $\lambda \simeq 1060$ Å at 1.9 K in the Y and Lu compounds. Eskildsen[20] has done the same measurements in the case of Tm borocarbide. He did also determine the internal B field for all three compounds, simply by measuring the distance between the flux quanta. In Y and Lu borocarbides, the internal field was found to be equal to the applied one, within experimental accuracy, whereas the internal field in the Tm compound was a factor of about 1.15 times the applied c-axis field. The experimental results of Eskildsen for the Tm compound are shown in Fig. 3.
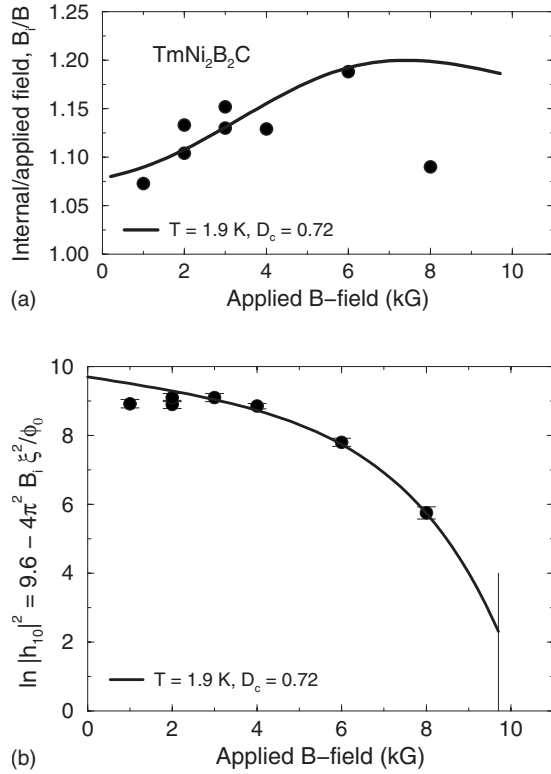
FIG. 3. The upper figure shows the ratio B_i/B as a function of the field B applied along the c axis of TmNi$_2$B$_2$C at 1.9 K. The points are the experimental results of Eskildsen (Ref. [20]) and the solid curve shows the calculated variation when the demagnetization factor $D_c=0.72$. The lower figure displays the field dependence of the logarithm of the flux-line-lattice form factor under the same conditions as in the upper figure. The neutron-diffraction results are from Ref. [20]. The solid line is the calculated result and the thin line indicates the upper critical field.

Except for the ratio at the highest field, the calculated results of H_i/H using Eq. (12) are in good agreement with the experimental values of B_i/B. The form factor is calculated by inserting

$$\xi^2(B) = \xi^2(0)\frac{H_{c2}}{H_{c2}^*} \tag{21}$$

into Eq. (20), and a reasonable fit is obtained when assuming $\xi(0)=89$ Å and $\ln|h_{10}|^2=9.6$ at zero field. The nonmagnetic upper critical field is $H_{c2}^0=H_{c2}=41.3$ kOe, which corresponds to the value of $\xi(0)=[\Phi_0/(2\pi H_{c2})]^{1/2}$ assumed in the fit. The upper critical field of the magnetic system, H_{c2}^*, is determined by the expression in Eq. (14) (at an arbitrary field) and decreases as the energy difference between the superconducting and the normal phase is increased, roughly proportional to the square of the field, and $\xi(B)/\xi(0)$ is found to be about 1.9 at the transition at 9.7 kOe. The zero-field value of the form factor indicates $\lambda\simeq 660$ Å, whereas the experimental value of about 8.9 corresponds to $\lambda\simeq 780$ Å. These results for λ and ξ agree with other determinations of these parameters and correspond to $\kappa\approx 8$, which is of the same order of magnitude as κ_0 used in the model. Due to the

Anderson-Suhl screening, the present theory predicts that the moments within the cores of the vortices are larger than those in the superconducting volume between the vortices, which difference reaches a value of about 0.8 μ_B per Tm ion close to the phase transition at 2 K [see Eq. (20)]. As first considered by Abrahamsen,[24] this excess magnetization of the cores is going to add to the scattering of the neutrons from the vortices. This effect is probably the explanation for the slight increase of the experimental form factor seen in Fig. 3, when the field increases from 0 to 3 kG. A more realistic interpretation of the field dependence of $|h_{10}|^2$ compared to the one presented here should include the effects of this additional scattering mechanism.

B. ErNi$_2$B$_2$C

The magnetic properties of the Er compound are well documented.[13,14,17,25,26] At low temperatures, the system is very hard to magnetize along the c axis, and within the ab plane it shows a four state clock behavior with the easy axes being along the a or b axes. Er borocarbide is superconducting below $T_c=11.2$ K and becomes antiferromagnetically ordered at $T_N=6$ K. The ordered moments are transversely polarized within the ab plane and the ordering wave vector is $Q\simeq(0.55,0,0)$. The hard c axis is not favorable for the quadrupolar ordering found in the easy c axis Tm system, and the quadrupolar phase is not occurring in this system,[19] and probably neither in the two other hard c-axis systems, Ho and Dy borocarbides.

The strong magnetic anisotropy implies that the periodicity of the antiferromagnetic structure becomes locked to the lattice periodicity, where the commensurable period depends on temperature and fields. At zero field, the commensurable structure has a period of 40 lattice planes, and the magnetic net moment is zero above $T_c\simeq 2.3$ K. Below this temperature, it becomes energetically favorable for the system to rearrange the structure into another 40-layer one, which has a small ferromagnetic component.[14] It is important to realize that the ferromagnetic transition in ErNi$_2$B$_2$C does not occur because of a ferromagnetic interaction, which interaction is actually strongly negative and is further reduced by the Anderson-Suhl screening. The ferromagnetic component has to be considered to be an accidental by-product of the low-temperature commensurable structure.

The effects of the magnetic Er ions on the upper critical fields of ErNi$_2$B$_2$C have been calculated using the model derived in Refs. [14] and [26] and the parameters in Table I. The results are shown in Fig. 4. In the c-axis case, the magnetic moment induced by the field is very small, implying that the Anderson-Suhl screening has nearly no consequences. Below the second-order transition at about 6 K, the rapid increase of the ordered moment results in a steep reduction of the upper critical field. When the field is applied in the ab plane, along [100] or [110], the reduction is appreciable in the paramagnetic phase. Because of the large moment induced by the field, the transition to the antiferromagnetic phase is a first-order one, but the amplitude of the oscillating moments stays relatively small below the transition. Correspondingly, the theory predicts a sharp, but small jump in the upper critical
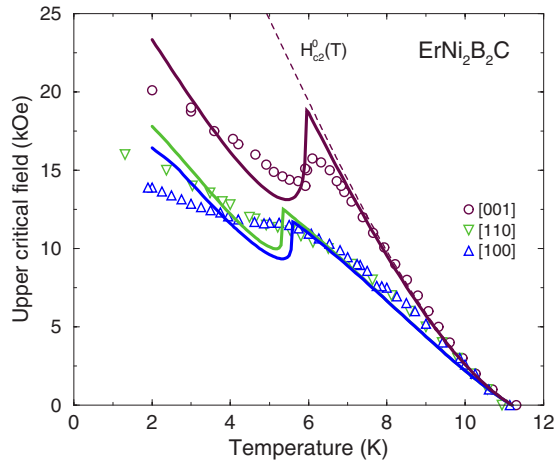
FIG. 4. (Color online) Upper critical field in $ErNi_2B_2C$. The experimental results are from Ref. 27. The dashed line denotes the assumed value of $H_{c2}^0(T)$ when the field is along [001]. The solid lines are the calculated results using $D_c = 0.6$.

FIG. 5. (Color online) Upper critical field in $HoNi_2B_2C$. The experimental results are from Ref. 32. The thin dashed line denotes the assumed c-axis value of $H_{c2}^0(T)$. The solid lines, and the dashed extrapolations, are the calculated results using $D_c = 0.64$. Below about 3 K, when the field is along [100] or [110], the calculated upper critical fields coincide with the metamagnetic transition fields of about 5 kOe, as indicated by the thick dashed lines.

field at the transition to the antiferromagnetic phase. The experimental upper critical field does not show a jump, but only a change of slope at the transition. The reason might be that the experimental transition is smeared out due to the inhomogeneities introduced by the relatively large demagnetization field (~ 1 kOe).

C. $HoNi_2B_2C$

The magnetic properties of Ho borocarbide are complex.[17,28–31] The magnetic anisotropy of the Ho system is similar to that of the Er system except that the easy axes are now the $\langle 110 \rangle$ directions. As discussed, for instance, by Lynn *et al.*[17] there is a competition between three different antiferromagnetic structures. The main transition is observed at about 6 K, and below this temperature the ordered phase is a simple commensurable antiferromagnet with $Q = (0, 0, 1)$ and the moments along a [110] direction. Between 6 and 8 K, the diffraction experiments show small scattering intensities at $(0,0,0.91)$ and at $(0.55,0,0)$. Because of the weak intensities, these extra features do not have much influence on the present calculations and are neglected. For the same reason as in the Er compound, the Anderson-Suhl screening has no influences on the calculated upper critical field, when applying the field along the hard c axis, instead the superzone effects are strong below the second-order phase transition, which is assumed to occur at 6 K (see Fig. 5). When the field is applied in the ab plane, the Anderson-Suhl screening is of some importance, and in the antiferromagnetic phase the superzone effects are appreciable but less pronounced than in the c-axis case. The experiments show a number of metamagnetic spin-flop transitions.[28,29] The lowest-field transition occurs at about 5 kOe, both when the field is applied along [100] or along [110], and the uniform moment makes a jump from nearly zero to about 3 μ_B/Ho. We have not developed a model for describing these transitions with sufficient accuracy (see instead Ref. 30); however, the calculations show that the uniform moment above the transition is sufficiently
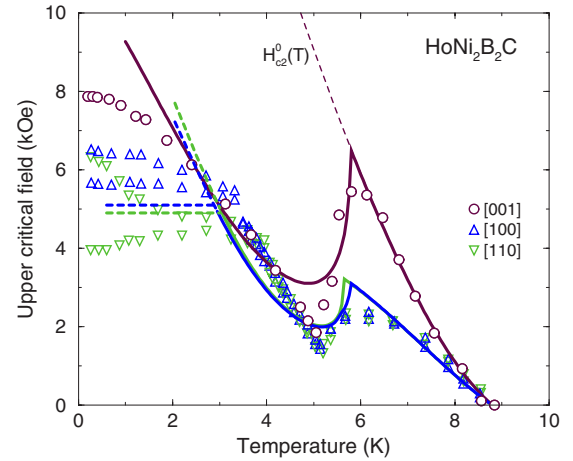
large so that the superconducting state is quenched because of the Anderson-Suhl screening effect. Hence, the present model predicts that the upper critical field along [100] or [110] coincides with the field determining the lowest metamagnetic transition, when the temperature is below about 3 K. Experimentally, this field is about 5 kOe roughly independent of temperature, as assumed in Fig. 5, but hysteresis effects are important.

D. $DyNi_2B_2C$

The Dy compound shares many of its magnetic properties with Ho borocarbide.[17,33,34] The easy axes are the $\langle 110 \rangle$ directions, and the magnetically ordered phase has the ordering wave vector $Q = (0, 0, 1)$. The main difference is that the superconducting phase is only stable within the antiferromagnetic part of the phase diagram. The superconducting transition at $T_c \simeq 6.5$ K is below $T_N = 10.6$ K and the lowest-field metamagnetic transition in this system occurs at an ab field which is larger than the upper critical field, at increasing-field conditions. Below the metamagnetic transition, the magnetization induced by the applied field is practically zero, and the Anderson-Suhl screening has no influence on the phase diagram. Instead, the superzone gaps are important and they produce nearly the same effects whether the field is applied parallel or perpendicular to the c axis; hence, the anisotropy of the calculated upper critical fields is alone due to the assumed 15% difference between the nonmagnetic H_{c2}^0 values. This is an accordance with the experimental results, as shown in Fig. 6. Utilizing decreasing-field conditions, Peng *et al.*[33] succeeded in maintaining a metastable spin-flop phase down below the critical fields shown in Fig. 6, at the lowest temperatures. In this situation, the upper critical field was found to coincide with the field of the spin-flop transition. This is entirely equivalent with the low-temperature behavior of the Ho compound.
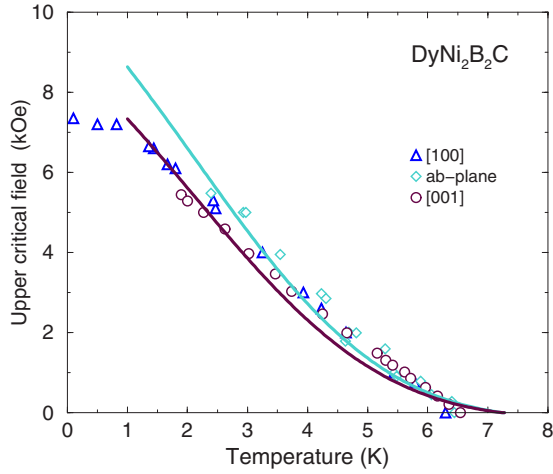
FIG. 6. (Color online) Upper critical field in DyNi$_2$B$_2$C. The experimental [100] results are from Ref. 33 and are those obtained for increasing fields. The $H \parallel c$ and $H \perp c$ data are from Ref. 34. The solid lines are the calculated results.

IV. CONCLUSION

The Tm, Er, Ho, and By borocarbides have the unique property of allowing superconductivity to exist simultaneously with an antiferromagnetic ordering of the rare-earth moments. The 4f moments of the rare-earth ions do certainly interact with the superconducting electrons, and, probably, the largest part of the interaction between the 4f moments is the RKKY interaction, which involves the conduction electrons in a direct fashion. The acting of the RKKY interaction implies not only that the superconducting Cooper pairs are scattered by magnetic fluctuations but also that the RKKY interaction is partly screened in the superconducting phase at long wavelengths, and that the antiferromagnetic ordering creates energy gaps at the Fermi surface, which superzone gaps reduce the effective density of states determining the superconducting order parameter. The main purpose of the present paper is to convince that the two phenomena associated with the RKKY interaction, the Anderson-Suhl screening of the ferromagnetic RKKY component and the superzone gaps produced by the antiferromagnetic ordering, are essential for understanding the properties of these unusual rare-earth compounds.

Effectively, we have used four fitting parameters for each of the compounds, T_c^0, $H_{c2}^0(0)$, $\kappa_0^2 \Delta \mathcal{J}$, and d_Q. The two first ones and κ_0 establish the superconducting properties of the "nonmagnetic" compound. $[H_{c2}^0(0)]^{1/2}$ and T_c^0 scale approximately in the same fashion, and κ_0 is chosen to obey the same scaling, from one borocarbide to the next including the Y and Lu compounds. Some of the differences are due to real changes of the electronic properties, but we also expect that they, effectively, reflect influences of the magnetic fluctuations. In order to access the relative importance of these fluctuations, we have calculated the paramagnetic low-energy response (below $\sim$0.4 meV) of the rare-earth moments in the different cases. The weakest fluctuations are found in the Tm system, and here they are reduced by a factor of 3 (Ising-like) compared to the free ion. The strongest ones, i.e., fluc-

tuations as large as for a free ion, are found in the Ho and Dy compounds, leaving Er borocarbide as an intermediate case. The conditions in the case of Dy are special, as the superconducting phase is only appearing at temperatures where the low-energy magnetic fluctuations are totally removed, because of the antiferromagnetic ordering. This may partly explain why the superconducting model parameters of Dy are those in Table I that are closest to the Y and Lu values. Incidentally, the reentrant behavior suggested by the model parameters of Dy, because $T_c^0 > T_N$, is assumed to be prevented by the strong fluctuations of the Dy moments close to and above T_N. Leaving Dy as a special case, then Ho borocarbide should be the one that is most influenced by magnetic fluctuations, even more so when considering that the interaction is also expected to be proportional to $(g-1)^2$. This is consistent with that Ho borocarbide is the weakest nonmagnetic superconductor according to Table I.

In the present model, the interactions between the magnetic rare-earth ions and the superconducting electrons are determined in terms of the two parameters $\Delta \mathcal{J}$ and d_Q. In the construction of the theory, we have been forced to make a number of quite drastic simplifying assumptions. For instance, we have assumed a simple isotropic superconducting gap Δ although indications of gap anisotropy have been observed, in particular, in the case of the nonmagnetic borocarbides,[35] as reviewed recently by Gupta.[3] The possible presence of nodes in Δ is going to change the temperature variation of $\chi_s(0)/\chi(0)$ shown in Fig. 1, especially in the zero-temperature limit, but even major modifications of the relative variation of this ratio between 0 and 1 would not be of much importance for the present analysis. The theory accounts convincingly for many particulars of the experimental observations. The model rightly predicts that the anisotropy of the upper critical field should have the opposite sign in the Tm compound compared to the hard c-axis systems Er and Ho. It is important to realize that if the magnetic fluctuations should be dominating the systems, the sign of the anisotropy would be the opposite: the fluctuations are most effectively reduced when the field is applied along an easy axis. Actually, in the cases of the Er, Ho, and Dy borocarbides, the field along the hard c axis has nearly no consequences for the magnetic systems within the superconducting phase. In this situation, the superzone effect is the only one left, which, in addition, is stronger here than in the easy-axis cases, simply because the hard-axis field does not reduce the antiferromagnetic order parameter. The superzone gaps are much largest in the Dy case, where the model indicates that the effective density of states at the Fermi surface is being reduced by up to 20%. The ordering wave vector $\boldsymbol{Q}$ is the same in the Ho and in the Dy system, and d_Q is smaller in Ho than in the Dy case, although the difference is somewhat larger than expected from the $(g-1)$ scaling. The superzone effect is capable of accounting for most part of the near-to-reentrant behavior of the Ho compound, but it is also clear that the magnetic fluctuations are important for this particular phenomenon, which fluctuations may be enhanced by the competition between the three different antiferromagnetic order parameters close to T_N. The interaction parameter d_Q is larger in the Tm compound than expected from the $(g-1)$ scaling, which may indicate that the unique quadrupolar or-

dering of this system leads to important deformations of the Fermi surface. Except for the case of Tm, d_Q scales roughly like $(g-1)$.

The Anderson-Suhl coupling parameter $\Delta\mathcal{J}$ is expected to be proportional to $(g-1)^2$, which is not the case. This is a somewhat disturbing result; however, this parameter is determined by finer details of the electronic bands near the Fermi surface, which do not necessarily depend in a systematic way on the rare-earth system considered. The importance of the individual electronic properties of the different rare-earth systems may be exemplified by comparing the exchange interaction determined in Tm and in Er borocarbide: In the Tm system, $\mathcal{J}(\mathbf{0})$ is close to the maximum value $\mathcal{J}(\mathbf{Q}_F)$, whereas this parameter is strongly negative in the Er system. In the Er system, $\mathcal{J}(\mathbf{q})$ has a sharp maximum at the ordering wave vector $\mathbf{q} \simeq \mathbf{Q}_A$, whereas $\mathcal{J}(\mathbf{Q}_A)$ is negative in the Tm system.[14,19]

One of the special phenomena in these magnetic superconductors is that the uniform polarization of the magnetic moments adds to the internal magnetic field [Eq. (12)]. This effect is straightforwardly included in the model calculations. It is not insignificant, but it is neither of great importance since it only adds a few kilogauss, at maximum, to the internal field and thereby reducing the applied upper critical field with the same relatively small amount.

The analysis of, in particular, the Tm system leaves no doubt that the Anderson-Suhl screening is important, as already concluded by Nørgaard *et al.*[7] It explains why this system does order at the small wave vector $\mathbf{Q}_F$ instead of zero—and why the antiferromagnetic phase stays stable up to a *c*-axis field of the order of 8 kOe—and also explains the increase of the radius of the vortices with field and the change of slope in the magnetization curve when the system becomes normal. For completeness, we point out that the clear indications of the Anderson-Suhl screening effects rule out the possibility that the rare-earth borocarbides are triplet superconductors; they must be singlet BCS superconductors, since the formation of the singlet ground state is the basic condition for the presence of screening.

Note added in Proof. Recently we learned of a detailed analysis of the effects of the paramagnetic moment on the vortices in Tm brorcarbide by L. DeBeer-Schmitt *et al.* [36]

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