Ferromagnetic Superconductors: A Vortex Phase in Ternary Rare-Earth Compounds

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It is shown that the generalized Ginzburg-Landau free-energy functional of Blount and Varma admits self-consistent solutions with quantized-flux vortices, magnetized in a region about the cores. There is a temperature range where the new phase has a lower free energy than either the pure superconducting or pure ferromagnetic phases; it represents true coexistence of ferromagnetism and superconductivity. The main features of the specific heat and magnetic properties of some rare-earth ternary compounds can be explained qualitatively.

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In the search for coexistence of superconductivity and ferromagnetism two classes of ternary rare-earth compounds are especially interesting: the rare-earth molybdenum chalcogenides[1] and the the rare-earth rhodium borides[2]. For example, the compound $ErRh_4B_4$ becomes superconducting (Type II) at a temperature $T_{c1} = 8.5$ K, but ceases to be superconducting—and becomes magnetically ordered—at $T_{c2} = 0.92$ K.

Most theoretical studies of these systems[3] have emphasized the Cooper-pair-breaking role of spin-spin coupling. This manifest itself (a) via spin-flip scattering and (b) via conduction-electron polarization. But recently several authors[4] have stressed the role of the direct coupling via the magnetic field. In particular BV[4] suggest that this is the dominant mechanism, and have proposed a generalized Ginzburg-Landau (GL) model; they introduce the free-energy functional

$$F = \int d^3r \{ \tfrac{1}{2}\alpha_s |\psi|^2 + \tfrac{1}{4}\beta_s |\psi|^4 + (\hbar^2/2m) |[-i\nabla - (2e/\hbar c)\vec{A}]\psi|^2 + (8\pi)^{-1}H^2 + \tfrac{1}{2}\alpha_m |M|^2 + \tfrac{1}{4}\beta_m |M|^4 + \tfrac{1}{2}\Gamma |\nabla\vec{M}|^2 + \tfrac{1}{2}(\eta_1 |M|^2 + \eta_2 |\nabla\vec{M}|^2) |\psi|^2 \}, \quad (1)$$

where $\vec{H} = \vec{B} - 4\pi\vec{M}$ and $\vec{B} = \nabla \times \vec{A}$, and where our definition of the dimensionless coefficient α_m differs from that of BV: $\alpha_m = \alpha_{BV} - 4\pi$. In the absence of superconductivity our α_m is directly related to the Curie temperature T_m in the same way as α_s is related to T_{c1}:

$$\alpha_s = \alpha_{s0}(1 - T/T_{c1}), \quad \alpha_m = \alpha_{m0}(1 - T/T_m). \quad (2)$$

The terms containing the coefficients Γ, η_1, and η_2 are, respectively, the contributions from the stiffness of the magnetization $\vec{M}$, the polarization of conduction electrons, and the spin-flip scattering. We believe, with BV, that the terms in η_1 and n_2 are of secondary importance; henceforth, for simplicity, we shall ignore them. We differ from BV in that we do not require Γ in order to stabilize the magnetic/superconducting state; in the BV screw structure Γ plays an essential role, despite its smallness ($\Gamma/\lambda \sim 10^{-2}$). In an analogy to the mixed state of a regular Type-II superconductor we seek a *self-consistent* vortexlike state of coexistence between superconductivity and ferromagnetism. We find such a solution: a vortex structure with ψ vanishing along the vortex cores, and with spontaneous magnetization around the cores. Near the center of a vortex, ψ rises rapidly to its bulk value with a healing length $\xi = \lambda/\kappa$. Magnetic order is however not confined to the region where ψ is small; its attenuation is characterized by a scale length longer even than the London length λ. The spontaneous magnetization plays a role analogous to external field in the Abrikosov problem.

Note that even at the vortex core $\vec{H}$ does not vanish; in other words, the magnetization does not attain the value which would be optimal in the absence of superconductivity. The vortex state is a compromise: Both the superconducting order and the magnetic order fall short of their individual optimum values. The new phase is characterized by the true coexistence of superconducting and ferromagnetic order, such that the *total* free energy is minimized.

We define dimensionless units by

$$\vec{x} = \gamma\vec{r}, \quad \vec{h} = \vec{H}/\sqrt{2}H_c\lambda, \quad \vec{a} = \vec{A}/\sqrt{2}H_c\lambda, \quad \vec{b} = \vec{B}/\sqrt{2}H_c, \quad \vec{m} = \vec{M}/M_0, \quad f = \psi/\psi_0, \quad (3)$$

where

$$\psi_0^2 = -\alpha_s/\beta_s, \quad \lambda^{-2} = 16\pi\psi_0^2/mc^2, \quad H_c^2 = 2\pi\alpha_s^2/\beta_s, \quad M_0 = -\alpha_m/\pi_m. \quad (4)$$

We also introduce the GL parameter κ, the ratio

ζ of magnetic to superconducting free-energy densities (at $T=0$), and two other dimensionless parameters ϵ and γ:

$$\kappa^2 = 8e^2\lambda H_c{}^2/\hbar^2c^2, \quad \zeta = \alpha_{m0}{}^2\beta_s/\alpha_{s0}{}^2\beta_m, \quad \gamma^2 = \frac{2\pi\zeta}{\alpha_{m0}}\frac{1-T/T_m}{1-T/T_{c1}}, \quad \epsilon = \frac{4\pi}{\alpha_{m0}\gamma}\frac{1}{1-T/T_m}. \tag{5}$$

To construct an explicit solution of the GL equations for a single quantized vortex, we assume axial symmetry about the z axis, and rewrite the free energy (1) in cylindrical polar coordinates (per centimeter):

$$F = \tfrac{1}{2}H_c{}^2\lambda^2\int_0^\infty dx\, x\{(1-T/T_{c1})^2[-f^2+\tfrac{1}{2}f^4+\kappa^{-2}f'^2+(Q^2/x^2)f^2+h^2]+\zeta(1-T/T_m)^2[-m^2+\tfrac{1}{2}m^4]\}. \tag{6}$$

Here we have assumed the single-flux-quantum condition $f(\vec{x}) = f(x)e^{i\theta}$ and have defined[5] $Q=\kappa^{-1} - xa$. Functional differentation of F with respect to f, m, and Q gives[6]

$$-\kappa^{-2}x^{-1}(d/dx)(xdf/dx)+x^{-2}Q^2f = f-f^3, \tag{7}$$

$$m^3 - m = \epsilon h, \tag{8}$$

and a second-order equation for Q which is conveniently decomposed into two first-order equations:

$$dh/dx = -x^{-1}Qf^2, \tag{9}$$

$$dQ/dx = -x(h+\gamma m). \tag{10}$$

Equations (9) and (10) are, respectively, the Maxwell equation curl $\vec{H} = (4\pi/c)\vec{J}$ and the definition $\vec{B} = \text{curl}\,\vec{A}$ in dimensionless units and with axial symmetry.

The system of Eqs. (7)–(10), contains, implicitly or explicitly, five material parameters: κ, T_{c1}, T_m, α_{m0}, and ζ. The method of solution will, necessarily, be numerical. We must therefore choose values for these parameters before we can proceed. In order to interpret the experimental results[2] for $ErRh_4B_4$, we shall choose $T_{c1}=10$ K, $T_m=1.5$ K, $\kappa=10$, $\alpha_{m0}=60$, and $\zeta=10$. To motivate this choice of α_{m0} and ζ, we note that at zero temperature M_0 satisfies $M_0(0) = (\alpha_{m0}/\beta_m)^{1/2} = \mu_I N_m$, where for Er the magnetic moment μ_I of a single ion is ~5 Bohr magnetons, and where N_m is the number of magnetic ions per unit volume. The unit cell is nearly cubic with side ~6 Å, and hence $N_m \sim 5\times10^{21}$ cm^{-3}. The magnetic GL coefficients also satisfy $\alpha_{m0}{}^2/4\beta_m = N_m kT_m$. Eliminating β_m, we find $\alpha_{m0}\sim 60$. The analogous relation for α_s, β_s is $\alpha_{s0}{}^2/4\beta_s = H_c{}^2/8\pi \simeq N_c kT_{c1}(kT_{c1}/\epsilon_F)$, where N_c, the number of conduction electrons per unit volume, is ~20 N_m. The factor $kT_{c1}/\epsilon_F \sim 10^{-3}$ reflects the fact that only electrons in a shell of thickness $\sim kT_{c1}$ close to the Fermi energy ϵ_F take part in the superconducting transition. Comparing these energies, we find $\zeta\sim 10$.

Note that the equations reduce to Abrikosov's when $\epsilon=\gamma=0$. We therefore expect a solution similar to Abrikosov's—and indeed we shall find such a solution. The dimensionless radius of the "nonsuperconducting" core is κ^{-1}, the magnetic induction b falls off with a characteristic length ~1, and the total flux is one quantum, $\Phi_0 = \pi\hbar c/e$. The magnetization is even more spread out than the induction; quite a small density of vortices will allow a fair degree of homogeneity of the magnetization.

The asymptotic solution is straightforward; assume that m is small, and neglect its cube in Eq. (8). Then the equations become identical[6] to Abrikosov's, but with a rescaled length unit: $x' = \lambda^{-1}r(1-\gamma\epsilon)^{-1/2}$. Provided[7] that $1-\epsilon\gamma>0$, we recover an Abrikosov-like vortex, with nearly exponential decay

$$m \sim -h \sim \kappa_0[x(1-\gamma\epsilon)^{1/2}]. \tag{11}$$

We have integrated the equations numerically for several temperatures. Figure 1 shows the behavior of f, m, h, and b, over the entire plane, for $T=0.75$ K. Note that at this temperature $(1-\gamma\epsilon)^{1/2}$ is quite small, and hence m is still about a third of its maximum value at $x=2$. In contrast, b is ~0 by $x=2$.

We must still show that this solution of the GL equations represents a *minimum* of the free energy. Substituting our solution back into (6), we find that there is a transition temperature $T_v \sim 0.9T_m$, below which F is negative, i.e., the vortex is stable.

As soon as one vortex is energetically favored, many will appear. Their equilibrium distance is determined by their interactions. When the vortices are well separated, the interaction energy is the integral of

$$(1-T/T_{c1})^2h_1h_2 - \zeta(1-T/T_m)^2m_1m_2, \tag{12}$$

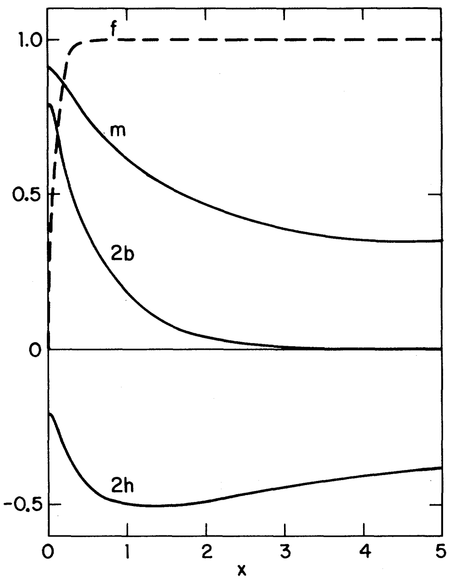

FIG. 1. Numerical solutions for a single vortex: $T = 0.75T_m$.

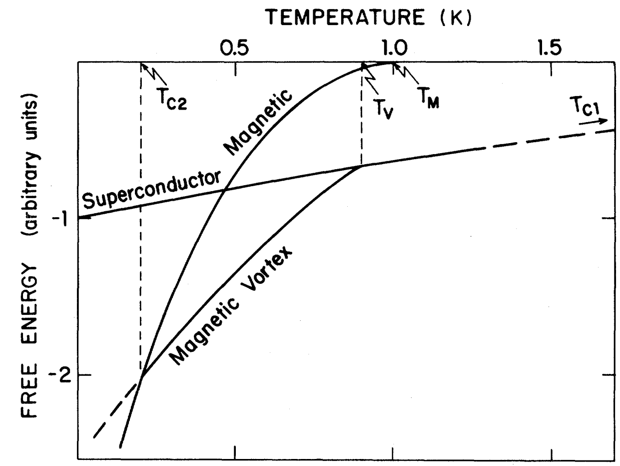

FIG. 2. Schematic diagram of free energy against temperature.

over the region where the "tails" of m and h overlap. For parallel vortices (12) is negative; i.e., magnetic vortices attract at large distances. At short distances they repel; the equilibrium distance at $T = 0.8T_m$ is $x_l \sim 2$. The temperature dependence of x_l is weak; the main influence of temperature is to change the value of M_0.

The transition at T_v is qualitatively like the Abrikosov transition at H_{c1}. But the attraction between vortices will lead, *prima facie*, to a rather weak first-order transition. In temperature units the latent heat will be roughly $T_m(1 - T_v/T_m)^2 \sim 10^{-2}$ K.

The lower critical temperature T_{c2} will be given by the intersection of the free-energy curves for the vortex phase and the pure ferromagnetic phase (see Fig. 2). T_{c2} will be characterized by the collapse of the superconducting order, as well as by a possible jump in the magnetization (since in the vortex phase m is in the range $\frac{1}{2} \lesssim m < 1$). The collapse of superconductivity will contribute $T_{c1}(1 - T_{c2}/T_{c1})(T_{c1}/\epsilon_F)(N_c/N_m) \sim 2\times 10^{-1}$ K to the latent heat. Thus, *even if there is no discontinuity in the magnetization,* the transition at T_{c2} is much stronger than that at T_v.

In contrast to the Abrikosov structure, the direction of the magnetic induction is not determined by external constraints. Indeed in our isotropic model, the direction of the vortices is arbitrary. The vortex structure is less regular than Abrikov's, because the orientational degrees of freedom are easily excited. This explains qualitatively the increase[2] in the specific heat of $ErRh_4B_4$ between 1.3 K, which we tentatively identify with T_v, and 0.93 K (T_{c2}). (Even real anisotropic materials have several distinct axes of easy magnetization).

Another experimental result[1] which our picture explains is the fall in the critical field H_{c2} as $T \to T_{c2} + 0$. The magnetization contributes to the magnetic field, so that near T_{c2} the induction B will attain the value H_{c2} when the applied external field is still well below H_{c2}.

It should be possible to detect the magnetic vortex structure in neutron-scattering experiments. The distance between vortices is ~100 Å, quite similar to the Abrikosov lattice. It may already have been seen: Moncton's[8] small-angle neutron-scattering results are strikingly similar to those of the Saclay group on Nb in the mixed phase.[9]

To conclude, we have demonstrated the possibility of a vortex phase which is both superconducting and ferromagnetic. We should remark that in principle the vortex phase which is both superconducting and ferromagnetic. We should remark that in principle the vortex phase could persist down to zero temperature for suitable values of the parameters.

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[2]M. B. Maple, J. Phys. C 6, 1374 (1978); B. T. Matthias, E. Corenzwit, J. M. Vandenberg, and H. E. Barz, Proc. Nat. Acad. Sci. U. S. A., 74, 1334 (1977).

[3]See, for example, C. A. Balseiro and L. M. Falicov, Phys. Rev. B 19, 2548 (1979); S. Maekawa and M. Tachiki, Phys. Rev. B 18, 4688 (1978).

[4]E. I. Blount and C. M. Varma, Phys. Rev. Lett. 42, 1079 (1979) (hereafter BV), and 43, 1843(E) (1979); U. Krey, Int. J. Magn. 3, 65 (1972), and 4, 153 (1973); M. V. Jarić and M. Belić, Phys. Rev. Lett. 42, 1015 (1979); M. Tachiki, H. Matsumoto, and H. Umezawa, Phys. Rev. B 20, 1915 (1979). Krey was probably the first to propose the direct interaction, and he discussed the vortex structure (in an external field). Jarić and Belić have suggested the possible existence of a mixed state. Tachiki *et al.* have limited their discussions to the region $T > T_m$. However, a paper by M. Tachiki, H. Matsumoto, T. Koyama, and H. Umezawa (to be published) was received by us after submission of this paper. There appears to be some overlap between their results and ours. Varma has referred to the possibility of a vortex phase at the Proceedings of the Third Conference on Superconductivity in *d*- and *f*-Band Metals, La Jolla, California, June 1979 (to be published).

[5]More generally we can have multiply quantized vortices, with $f(x) = f(x)e^{-i\theta n}$ for integer n. Then $Q = n\kappa^{-1} - xa$.

[6]Cf. A. A. Abrikosov, Zh. Eksp. Teor. Fiz. 32, 1442 (1957) [Sov. Phys. JETP 5, 1174 (1957)]. Eq. (8) is Abrikosov's Eq. (26) and our (10) and (11) reduce to Abrikosov's (27) when $m = 0$.

[7]Very close to T_m, $1-\gamma\epsilon$ is certainly less than 0, so that instead of (12) we shall have $m \sim -h \sim J_0[(\gamma\epsilon - 1)^{1/2}x]$. However, the magnetic stiffness parameter Γ will tend to damp out these oscillations. The residual oscillations should have little effect.

[8]D. E. Moncton, J. Appl. Phys. 50, 1880 (1979), Fig. 5.

[9]D. Saint-James, G. Sarma, and E. J. Thomas, *Type II Superconductivity* (Pergamon, Oxford, 1969), p. 72, Fig. 3.10.

Neutron Scattering Study of Spin Dynamics in $CsCoCl_3$

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Neutron profiles of spin waves in the 1*d* Ising-type antiferromagnet $CsCoCl_3$ were reexamined. Instead of well-defined magnon peaks, the spin-wave spectrum consists of a band of magnon states arising from the motion of domain-wall pairs. At the magnetic zone center, the energy of this band extends from $2J(1-2\epsilon)$ to $2J(1+2\epsilon)$, whereas at the zone boundary a sharp resolution-limited peak at $\omega \sim 2J$ is observed.

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Statics and dynamics of spins in nearly ideal one-dimensional (1D) magnetic materials have been widely studied in the past several years.[1] The magnetic compound $CsCoCl_3$ has been found to behave as an antiferromagnetic chain of spins with $S=\frac{1}{2}$.[2] Tellenbach[3] and recently Yoshizawa and Hirakawa[4] have measured the spin-wave spectrum of $CsCoCl_3$ and found it to conform to the Ising-type Hamiltonian

$$\mathcal{H} = -2J\sum_i [S_i^z S_{i+1}^z + \epsilon(S_i^x S_{i+1}^x + S_i^y S_{i+1}^y)] = \mathcal{H}_{zz} + \mathcal{H}_{xy} \tag{1}$$

with $J = 6.5 \pm 0.5$ meV and $\epsilon = 0.094$.[3] These values of J and ϵ were arrived at by use of the spin-wave dispersion for anisotropic antiferromagnetic chains derived by des Cloizeaux and Gaudin (dCG)[5] and Tellenbach.[3] In the experiments of Refs. 3 and 4, the peak position of the spin waves could be obtained with certainty, but the neutron intensity and resolution were not sufficient to permit a detailed investigation of the line shape of neutron profiles as a function of Q and temperature.

Very recently, starting from the Ising Hamiltonian $\mathcal{H}_{zz}$ and treating $\mathcal{H}_{xy}$ as a perturbation, Ishimura and Shiba[6] have calculated the dynamical correlation functions $S_{xx}(Q,\omega)$ and $S_{zz}(Q,\omega)$ for $S=\frac{1}{2}$, 1D, Ising-type antiferromagnets given by the Hamiltonian (1). Their calculations show that the spin-wave dispersion in such systems is not an isolated branch calculated by dCG, but that it consists of a continuum of excited states. The spin-wave excitation spectrum calculated by Ishimura and Shiba thus looks like a band of states with energy extending from $2J(1-2\epsilon)$ to $2J(1+2\epsilon)$ at the zone center, whereas at the zone boundary

