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Influence of strain on the coexistence state of ferromagnetic superconductors

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The coexistence of sinusoidal magnetic order and ferromagnetism in the superconducting phase of $ErRh_4B_4$ is explained by the existence of an inhomogeneous strain which locally creates additional magnetocrystalline anisotropy, enhancing the ferromagnetic-phase-transition temperature more than the superconducting-sinusoidal-phase-transition temperature.

Many experimental and theoretical studies have investigated the coexistence state composed of sinusoidal magnetic order and ferrromagnetism which occurs in a narrow temperature range just above the reentrant phase transition temperature T_{c2} in $ErRh_4B_4$.[1,2] In recent neutron diffraction experiments using single crystals of $ErRh_4B_4$, intense ferromagnetic lines were observed along with satellite lines arising from the sinusoidal magnetic order in the temperature range of 0.7–1.1 K.[3] Magnetic measurements show that the susceptibility along the $\vec{a}$ axis gradually changes from the perfect diamagnetic value to that of the normal ferromagnetic state with decreasing temperature from 1.1 to 0.7 K.[4] The peaks of the Fraunhofer patterns in the maximum Josephson current shift and split in the same temperature range.[5] The most plausible explanation of these experimental results seems to be a coexistence of normal ferromagnetic domains and superconducting sinusoidal domains with the ferromagnetic volume continuously growing as the temperature is lowered below 1.1 K. The analysis of the linewidths of neutron diffraction indicates that the domain size is larger than 10000 Å.[6] All experimental results are reproducible with temperature, suggesting that the same stable pattern of normal and superconducting domains is established on each cooling cycle.

An important unresolved question raised by the interpretation of the data in terms of separate normal ferromagnetic and superconducting sinusoidal phases is the mechanism which stabilizes the composite state. In general, the entire sample is expected to convert to the phase which has the lower free energy. In this paper we discuss a mechanism which naturally explains the simultaneous existence of the two phases and the continuous increase in volume of the normal ferromagnetic phase as the temperature is lowered. The basic feature of the mechanism is the existence of inhomogeneous strain in the sample which locally increases the magnetic anisotropy and enhances the transition temperature of both the normal ferromagnetic phase and the superconducting sinusoidal phase.[7] We show that this additional magnetic anisotropy favors the ferromagnetic phase, so that the superconducting sinusoidal phase is not stable at any temperature when the strain is large enough. The strained regions of the sample then produce the ferromagnetic domains, while the superconducting sinusoidal domains occur in the relatively unstrained regions. We estimate the additional magnetic anisotropy needed to exclude the superconducting sinusoidal phase, and propose experiments to test the applicability of this mechanism.

It has been shown that magnetic anisotropy due to crystalline fields strongly enhances the Curie temperature T_m in the MRh_4B_4 series (with M a rare-earth metal) and that the enhancement explains much of the systematic change of T_m in these compounds. Previous work has shown that B_2^0 is the dominant term in the crystal-field Hamiltonian producing strong uniaxial magnetic anisotropy.[8] For $ErRh_{44}B_4$ this term tends to limit the spin direction to the basal plane, reducing the effective dimensionality of spin space from 3 toward 2. While additional crystal-field terms allowed by the symmetry of the Er site introduce anisotropy in the basal plane, the initial susceptibility (which is important in determining T_m) remains isotropic. Here we consider the effect of strains that remove this isotropy in the basal plane, further reducing the effective spin dimensionality from 2 toward 1. The effect of this reduction in dimensionality on the ferromagnetic transition temperature is illustrated by noting that an isotropic classical mean-field model gives T_m proportional to $J^2/3$, $J^2/2$, or J^2 for spin dimensionalities of 3, 2, or 1, respectively. Thus the reduction from 2 to 1 dimensions can enhance T_m even more than the reduction from 3 to 2 dimensions.

We can estimate the influence of strain effects on T_m within a crystal-field model. Because of the crystallographic point symmetry of the Er ions, the crystal-field Hamiltonian contains five parameters $(B_2^0, B_4^0, B_4^4, B_6^0, B_6^4)$. With proper values of these parameters, one can describe the experimental results for the temperature dependence and the magnetic field dependence of the magnetization, the paramagnetic susceptibility, and the Schottky anomalies in the specific heat due to crystal-field splitting.[9] Strain along the a axis will cause additional crystal-field parameters B_2^2, B_4^2, and B_6^2 to be significant. Within a molecular-field model, the ferromagnetic transition temperature T_m can be found by solving

$$\chi_0(T_m) = \lambda^{-1} , \tag{1}$$

where $\chi_0(T)$ is the paramagnetic susceptibility, including crystal-field effects, and λ is the molecular-field constant. Using previously determined crystal-field parameters, and assuming B_4^2 and B_6^2 to be negligible, we have calculated T_m as a function of B_2^2. The result, shown in Fig. 1, indicates that a relatively small value of $B_2^2/B_2^0 = 0.01$ shifts T_m to 1.3 K, relative to an assumed value of 1 K for $B_2^2 = 0$. A crude nearest-neighbor point-charge calculation indicates that strain parameters of this size can be obtained by changing one of the basal-plane lattice parameters on the order of 0.005 Å.

Near T_m, the crystalline-field anisotropy energy due to

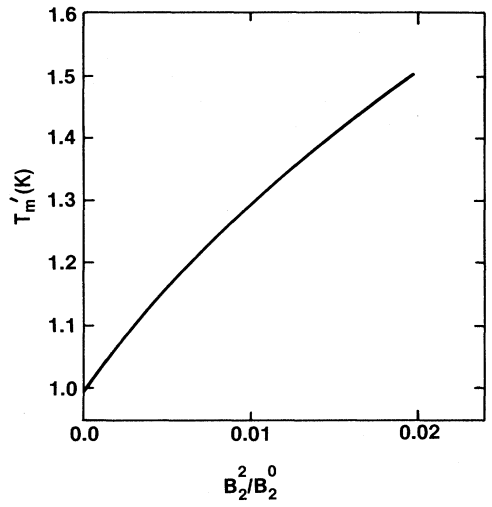

FIG. 1. Ferromagnetic transition temperature T_m as a function of B_2^2/B_2^0. T_m was calculated from Eq. (1) where χ_0 is the easy axis susceptibility in the crystal-field model with the parameters of Ref. 9, augmented with the values of B_2^2 shown in the figure. The mean-field constant λ was adjusted to give $T_m = 1$ K when $B_2^2 = 0$.

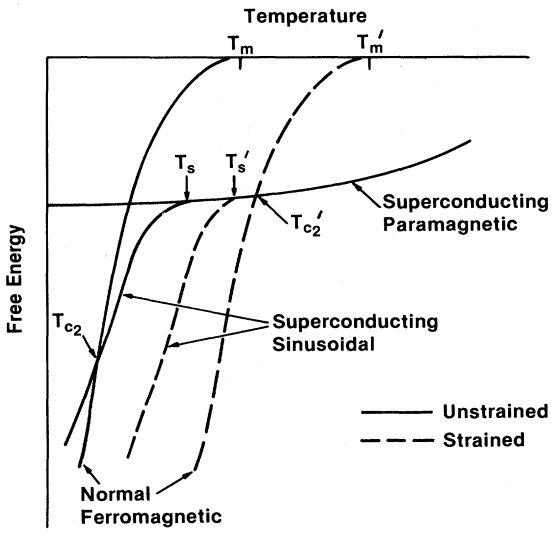

FIG. 2. Proposed free-energy diagram for ferromagnetic superconductors. The solid lines represent unstrained regions of the crystal, where the sample transforms from superconducting paramagnetic to superconducting sinusoidal to normal ferromagnetic. The dashed lines correspond to strained regions of the crystal, where additional magnetic anisotropy enhances both T_m and T_s, such that the sample transforms from superconducting paramagnetic to normal ferromagnetic with no intervening superconducting sinusoidal state.

internal strain can be written as

$$-A \sum_i m_i^2(T) \ , \tag{2}$$

where A is a constant related to the crystal field of Er and $m_i(T)$ is the magnetic moment of Er at the ith lattice site. In the ferromagnetic state, the expression (2) reduces to

$$-ANm^2(T) \ , \tag{3}$$

where $m(T)$ is the magnetization per Er ion and N is the number of Er ions per unit volume. In the sinusoidal state with wave number $\vec{Q}$, the expression (2) becomes

$$-Am'^2(T) \sum_i \sin^2(\vec{Q}\cdot\vec{R}_i) = -ANm'^2(T)/2 \ . \tag{4}$$

Since the amplitude of the sinusoidal magnetization $m'(T)$ is slightly smaller than $m(T)$, the energy (2) is more than twice the energy (3). Therefore the presence of strain enhances the magnetic transition temperature of the ferromagnetic state much more than that of the sinusoidal state.

We now combine the above comments with a free-energy discussion to provide a model of the coexistence state. In Fig. 2, the solid lines show schematically the situation for the unstrained regions. As the temperature is lowered the crystal transforms from the superconducting paramagnetic state to the superconducting sinusoidal state at $T = T_s$ and finally to the normal ferromagnetic state at $T = T_{c2}$. The free energies in a strained sample are obtained by adding (3) to the energy of the normal ferromagnetic state and (4) to the energy of the superconducting sinusoidal state. The result is shown schematically in Fig. 2 by dashed curves. Because of the large strain-induced anisotropy energy for the ferromagnetic state relative to the sinusoidal state, the order in which the magnetic states occur is reversed. Now, as the temperature is lowered, the crystal transforms directly to the ferromagnetic state at $T_{c2'}$, before reaching the sinusoidal state at $T_{s'}$,

The present model considers the observed ferromagnetic domains in $ErRh_4B_4$ to arise from strained portions of the material, while the sinusoidal order occurs in relatively unstrained regions. In perfectly strain free material, ferromagnetism would not coexist with superconductivity. Ferromagnetism appears in the strain-free regions at T_{c2}, when the superconducting sinusoidal state disappears. Correspondingly, in the highly strained regions, the superconducting sinusoidal state does not occur at any temperature.

A distribution of strain in the sample will lead to a distribution of Curie temperatures and reentrant temperatures. As the temperature is lowered, a greater fraction of the sample finds itself below its local value of $T_{c2'}$, leading to a gradual conversion of the superconducting sinusoidal volume to ferromagnetic volume. This explains the unusual concave upward curvature of the ferromagnetic intensity in the neutron scattering results,[3] and the continuous change in the low-field susceptibility from the Meissner value at 1.2 K to the ferromagnetic value at 0.7 K.[4] Some unannealed powder samples of $ErRh_4B_4$ have been reported not to show the satellite lines.[10] This fact may be understood if those samples include large strains, as may be expected in powders.

We do not expect the strain-induced anisotropy to cause a pronounced effect in all reentrant superconductors. For $ErRh_4B_4$, the effect arises because the moments are constrained to be in the basal plane, resulting in an effective spin dimensionality of 2. Strain can then reduce this dimensionality to 1 and enhance T_m. However, for materials which have moments constrained along a single axis, no further reduction in dimensionality, and thus no further

enhancement in T_m, can occur. This is the case for $HoMo_6S_8$, where the moments lie along the unique [111] axis of the rhombohedral crystal.[11] This explains in a natural way the absence of simultaneous ferromagnetic and superconducting sinusoidal order in $HoMo_6S_8$.[12,13]

Finally, we would like to propose the following experiments that may directly check the present model: (1) measurements of the satellite intensities under stress along the $\vec{a}$ axis (from the present model, a decrease of these intensities is expected under stress); (2) measurements of the magnetization and susceptibility in the temperature range of the coexistence state as a function of applied stress; and (3) magnetostriction measurements in the basal plane to obtain information on the strain dependence of B_2^2.

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