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Effect of radiation damage on the coexistent antiferromagnetic superconductor $SmRh_4B_4$

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Radiation-damaged films of the coexistent antiferromagnetic superconductor $SmRh_4B_4$ have been studied. The upper critical field, superconducting and magnetic transition temperature, and resistivity ratio were determined as a function of dose. The anomalous dependence of the superconducting transition temperature on resistivity ratio found previously for "as-made" films was also found in the damaged films, indicating the anomaly is intrinsic to $SmRh_4B_4$. A disorder-enhanced-spin-flip model is proposed to explain this effect.

INTRODUCTION

Materials exhibiting both magnetism and superconductivity have received a great deal of attention in the past few years.¹ Since the effect of disorder on superconductivity in ordinary (nonmagnetic) superconductors has also been intensively studied,² it is of interest to investigate the effect of disorder on the coexistent magnetic superconductors, both in terms of magnetic and superconducting properties. The effect of disorder on the ferromagnetic superconductor ErRh₄B₄ was studied by Rowell, Dynes, and Schmidt,³ and the transition temperature T_c versus resistivity ratio r_R was found to obey a universal curve first pointed out by Poate, Testardi, Storm, and Augustyniak.⁴ Previously, an anomalous behavior of T_c vs r_R in "as-made" films of the coexistent antiferromagnetic superconductor, SmRh₄B₄, was reported.⁵ This Rapid Communication reports identical behavior in radiation-damaged films of SmRh₄B₄, which clearly shows this to be an intrinsic property of SmRh₄B₄ and not a fundamentally less interesting proximity effect with low T_c impurity phases in the films.⁵ We have also studied the superconducting upper critical fields $H_{c2}(T)$ and thus derived the antiferromagnetic ordering temperature T_N as a function of disorder.

EXPERIMENT

Two sputtered films of SmRh₄B₄ were prepared as described previously using a triode sputtering gun.⁵ Half of one film, No. 31-2, was exposed to a dose of $0.5 \times 10^{16} \alpha$ particles/cm² and then, after making measurements, to a dose of $0.15 \times 10^{16} \alpha$ particles/cm². The other half of the film was sequentially exposed to low doses of α particles (1.8 MeV). A second film No. 39, was sequentially damaged with protons (0.25 MeV). For both films, after each irradiation, r_R , T_c , and $H_{c2}(T)$ were measured.

The parallel critical fields $H_{c2}(T)$ for various dose levels in each sample are shown in Fig. 1. The antiferromagnetic ordering temperature T_N was found by locating the position of the "kink" in each curve (~ 0.85 K). A good correlation between T_N and T_c was found, as shown in Fig. 2, which also includes as-made films for comparison. If the line in Fig. 2 is extrapolated to lower transition temperature, T_c will fall below T_N at ~ 0.78 K, and the film will order magnetically before becoming superconducting. Evidence for $T_N > T_c$ is seen in the $H_c(T)$ for our most heavily damaged film ($T_c = 0.68$ K), since no kink is seen. Note that for disordered ErRh₄B₄ films, a decrease in the *ferromagnetic ordering temperature* with decreasing T_c was also seen.³

Our measurements of T_c vs r_R for radiation damaged $SmRh_4B_4$ as well as as-made films are shown in Fig. 3. The as-made and irradiated films behave similarly and clearly disagree with the universal behavior discussed above. On the other hand, our ErRh₄B₄ films, as well as those of others, follow the universal behavior. Note that these data, as well as $H_{c2}(T)$ (Fig. 1) for the damaged films, are much more regular than for the as-made films (Ref. 5). The scatter of as-made films⁵ is very likely due to the different amounts, distributions, and grain sizes of trace impurity phases in the various films. Although the decrease in T_c with dose is anomalous for SmRh₄B₄, the decrease in r_R with dose is similar to that in ErRh₄B₄ and LuRh₄B₄ (see Fig. 4). Therefore, since r_R is a measure of disorder, the anomalous behavior of SmRh₄B₄ must be due to an anomalously fast depression of T_c with disorder.



FIG. 1. Upper parallel critical fields as a function of temperature for two samples after various doses of radiation. All curves are for the sample exposed to α -particle damage, except the (+) curve which is for proton damage.

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FIG. 2. A plot of T_N vs T_c for "as-made" films and radiationdamaged films of SmRh₄B₄. The bulk value (see, e.g., Ref. 11) is shown for reference.



FIG. 3. The variation of T_c/T_{c0} with residual resistance ratio r_R for "as-made" and radiation-damaged films of SmRh₄B₄. Also shown are ErRh₄B₄ films made by us and in Ref. 3 (including radiation-damaged films) and our results for VN films. The universal behavior of A15 superconductors is indicated by the shaded region. The scale on the right refers only to SmRh₄B₄ films where T_{c0} is the transition temperature of the bulk material.



FIG. 4. The residual resistance ratio r_R and T_c/T_{c0} are shown as functions of α -particle dose for our SmRh₄B₄ film, and for ErRh₄B₄ and LuRh₄B₄ films from Ref. 3, where T_{c0} is the transition temperature of the undamaged film.

DISCUSSION

Measurements of T_c vs r_R for a large class of superconductors exhibit a universal behavior⁴ which is shown as the shaded area in Fig. 3. Materials as diverse as A15 compounds, niobium,⁶ ErRh₄B₄, and our recent results on vanadium nitride (VN) films exhibit such universal behavior. The samples include as-made films and films progressively damaged by various radiations³ (α particles, neutrons, etc.). Although the exact nature of the disorder is unknown, two theoretical concepts have been suggested as being responsible. In the first,⁷ it is presumed that the Fermi energy E_F lies near a peak in the density of electron states N(E), and damage lowers $N(E_F)$ and, hence, T_c through the usual BCS relations. More recently, a localization model⁸ has been suggested, from which the universality of the behavior is more readily understood.

Since SmRh₄B₄ does not follow the universal curve, there must be an additional mechanism, not included in, but not contradicting either of the two models proposed. Any attempt to explain why SmRh₄B₄ is anomalous must include a reason why similar ErRh₄B₄ samples are not. Two models were previously proposed which could explain why ErRh₄B₄ follows the universal T_c vs r_R curve, but as-made SmRh₄B₄ films do not.⁵ The two models required different types of 5372

disorder: a model relying on proximity effect with normal phase impurity grains, which become more numerous with disorder; and a disorder-enhanced-spin-flip-scattering model, which resulted from disorder within the superconducting grains and is thus an intrinsic property of the SmRh₄B₄ phase. Radiation damage can distinguish between these, since it causes disorder within the superconducting grains, but will not change the number or size of any normal phase grains. Thus, radiation damage will not only enhance the disorder-enhanced spin-flip scattering, but will actually weaken the proximity effect, since disorder will reduce the coherence length. For the proximity model, T_c should not decrease as radiation damaged increases, but should approach the universal curve. Instead the data follow the behavior of as-made films, so any proximity effect explanation can be ruled out for the nonuniversal behavior.

DISORDER ENHANCED SPIN-FLIP SCATTERING

It is well known that spin-flip scattering is much smaller in ordered $R \operatorname{Rh}_4 B_4$ compounds (R denotes rare earth) than in materials with a comparable density of random magnetic ions (superconductivity is usually completely destroyed for $\sim 1\%$ random magnetic impurities). The reason is that the conduction electron wave functions, which are associated with the RhB clusters, have very little overlap with the localized spin on the rare-earth ions.⁹ Such a small overlap must rely on a crystal with a reasonable degree of longrange order. Consequently, any disorder in this perfect lattice structure should increase the overlap, enhance the spin-flip scattering, and lower T_c . The effect of spin-flip scattering on T_c can be analyzed within the context of the Abrikosov-Gor'kov (AG) theory¹⁰ which predicts a universal behavior for the reduction of T_c with the pair-breaking parameter, α .

The AG theory also forms a basis for understanding the effect of disorder on T_c in these compounds. The universal AG curve predicts the dependence of T_c on α , so the dependence of α on disorder is required. As discussed in Ref. 5, the only parameter in α which can reasonably be expected to change with disorder is f; the exchange interaction parameter between the conduction electrons and the localized rare-earth spins. The small value of f must rely on a reasonable degree of long-range crystalline order. Thus f, and hence α , could change significantly even for mild disorder.

Since the exact nature of the disorder is unclear, we make an *ad hoc* assumption that the small overlap is confined to

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the exponential tail of the wave functions. Then, if a given amount of disorder (measured by r_R) corresponds to a greater overlap, the increase in f will be proportional to f. Thus, as shown in Ref. 5, the change in T_c with disorder is proportional to $\alpha(dT_c/d\alpha)$, where both α and $(dT_c/d\alpha)$ are determined from the universal AG curve. To evaluate this for ErRh₄B₄ and the various SmRh₄B₄ samples, T_{c0} is assumed to be the value (11.4 K) for nonmagnetic LuRh₄B₄. Thus, one finds that $\alpha(dT_c/d\alpha)$ and hence the T_c depression for SmRh₄B₄ samples is approximately 5–11 times greater than for ErRh₄B₄, where the range covers T_c values for SmRh₄B₄ between the bulk value $T_{cb} = 2.72$ K and $T_{cb}/2$. This is entirely consistent with the data in Fig. 3.

At first glance, the decrease in the measured T_N with increasing disorder (Fig. 2) seems to be contrary to the above model in which f increases with disorder. This is because simple models predict the magnetic ordering temperature, T_M , to be proportional to f^2 . The important influence that the lattice crystalline electric field (CEF) has on magnetic ordering has been demonstrated¹¹ by inconsistencies in the determination of \mathcal{J} from $(dT_c/dn)_{n\to 0}$ and of T_M for the RRh₄B₄ series. The large magnetic anisotropies found in many RRh₄B₄ compounds (associated with CEF) can enhance T_M by effectively lowering the degrees of freedom of the magnetic moments. Disorder may reduce (or at least randomize) this anisotropy and hence T_M . Thus, there can be two compensating disorder effects on T_M (or T_N in the case of SmRh₄B₄) due to anisotropy changes and f^2 from the above model. Therefore, disorder-enhanced spin-flip scattering cannot be ruled out because T_N decreases with disorder.

In conclusion, we have measured superconducting and magnetic properties of disordered SmRh₄B₄. An anomalous T_c vs r_R behavior was found, indicating a new disorder effect in SmRh₄B₄. This new effect is in addition to, rather than replacing previously proposed universal models.^{7,8} We have proposed a model which can explain this anomalous depression of T_c with disorder, based on an enhancement of the spin-flip scattering due to the decrease of long-range crystalline order.

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