Sputtered films of superconducting $SmRh_4B_4$

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Films of the coexistent antiferromagnetic superconductor $SmRh_4B_4$ have been made by sputtering. These films have been characterized by x-rays and superconducting critical temperatures T_c, fields, and currents. Our best films have residual resistance ratios, r_R, as high as or higher than for films of $ErRh_4B_4$ reported in the literature, and the highest T_c is 2.36 K. We find an anomalous dependence of T_c on r_R, and offer two possible explanations (proximity-effect and disorder-enhanced spin-flip scattering), as well as propose a radiation-damage experiment to distinguish between these. The antiferromagnetic ordering temperature is shown to decrease with film disorder as does the ferromagnetic ordering temperature in radiation-damaged $ErRh_4B_4$. We find an approximately 1000-Å-thick RhAl impurity phase at the sapphire-substrate/film interface which seems to result from a reaction with the substrate. The potential importance of RhAl and trace impurity phases on present measurements and planned tunneling experiments are discussed in detail. Finally, the full critical-field curves are presented along with an explanation of why the low-temperature values are independent of T_c.

I. INTRODUCTION

Materials exhibiting both magnetism and superconductivity offer the opportunity to study the coexistence of these ordered electron states. A great deal of experimental and theoretical research has resulted after the initial discovery of such materials.[1] In the case of coexistence of antiferromagnetic order ($T < T_N$) and superconductivity (for T below $T_c > T_N$), measurements of the upper critical field B_{c2} have revealed two characteristic behaviors in which B_{c2} either drops[2] or increases faster[3] as T falls below T_N. Theoretical models[4–9] have been presented which consider various combinations of four distinct mechanisms through which the antiferromagnetism affects the superconductivity. These models can explain either or both[6,9] of the above temperature dependences of B_{c2}, so that measurements of B_{c2} alone cannot differentiate between the models.

However, the superconducting condensation energy E_c, is a more direct measure of the effect of antiferromagnetism on superconductivity and can be used to distinguish between the various proposed models. Although magnetization measurements[10] are traditionally used to determine E_c, there are problems with flux pinning and determining the magnetization of the normal magnetic state;[11] electron tunneling[12] into superconducting films provides an alternative. The tunnel current at finite voltages can be related to E_c through the reduced density of electron states,[13] and Josephson tunneling[14] at zero voltage can give information on the superconducting pair density, which is closely related to E_c. This paper reports our progress in producing films of the antiferromagnetic superconductor $SmRh_4B_4$ for the purpose of tunneling studies. In characterizing these films, we have identified a RhAl phase resulting from a reaction with the sapphire substrate; discovered an anomalous dependence of T_c on the residual resistance ratio r_R; and have carefully investigated the temperature dependence of B_{c2}.

II. SAMPLE PREPARATION AND CHARACTERIZATION

The samples were sputtered films of $SmRh_4B_4$ which were characterized by x rays, superconducting transition temperature T_c, and residual resistance ratio r_R.

A. Sputter system and procedure

All films were made by dc sputtering using a Plasmax triode source which allowed independent manipulation of the discharge current and target potential (typically 400–800 V).[15] A stoichiometric target (0.6 in. diameter) was prepared in an induction furnace and had predominantly the correct phase. Approximately 45 films were deposited onto single-crystal sapphire substrates at temperatures from 800 °C to 1000 °C. Both orientations of substrate were used, i.e., c axis parallel (0°) and perpendicular (90°) to the film normal. Typical residual pressures in the chamber prior to sputtering were 2×10^{-7} Torr with the heater at high temperature. To assist in reducing the oxygen background during sputtering, high-purity argon (99.999%) was used in conjunction with a sublimation pump which deposited titanium onto a liquid-nitrogen-cooled copper plate. The argon gas was injected in the sputter gun and pumped through an orifice by the cryopump, resulting in a chamber pressure of about 6 mTorr of argon. Some samples were made at significantly higher pressures, up to 50 mTorr, either statically or with a dynamic flow, as above. For the lower pressure, a film of 4000 Å resulted after ~35 min of sputtering time.

After deposition the heater was immediately disconnected, and the sample cooled to ~250 °C in ~20 min, if the argon gas was pumped out. Some samples were cooled more quickly by leaving an argon pressure of 100 mTorr (~15 min) or by placing a liquid-nitrogen-cooled block in contact with the heater block (~11 min).

B. X-ray analysis

The simplest probe of the crystal phases present in our films is the conventional θ-2θ diffraction. However, thin films made on heated substrates usually show some degree of preferential orientation; thus the identification of minority phases is sometimes difficult and quantitative analysis is essentially impossible. To overcome this, it seems necessary to have a microprobe of the structure of individual grains using an analytical electron microscope perhaps employing convergent beam techniques.[16] Nonetheless, comparisons of x rays from film to film are useful, and tend to correlate roughly with the measured superconducting properties. Figure 1 shows x-ray scans for our target and one of our sputtered films using Cu Kα_1 radiation. Most of the expected peaks are observed although neither scan is representative of the calculated powder pattern[17] of YRh_4B_4, but in addition the relative intensities vary considerably from film to film. These results along with T_c and r_R are summarized for several representative samples in Table I. The lattice is slightly expanded relative to the bulk polycrystal values reported[17]

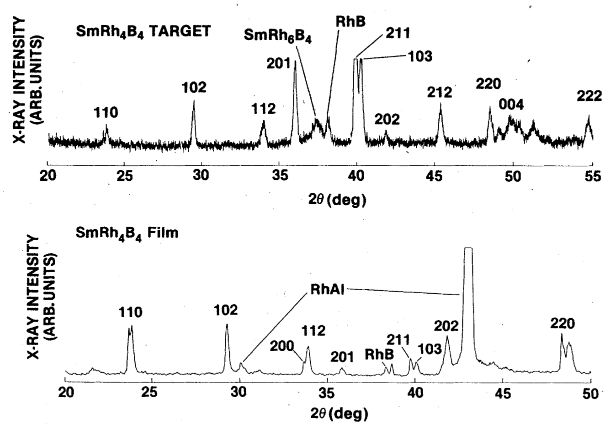

FIG. 1. X-ray diffraction patterns for the sputtering target and a representative sputtered film. The correct phase peaks ($SmRh_4B_4$) are indexed in both cases while impurity phases are labeled by stoichiometry. See text for discussion of RhAl in the films.

for $SmRh_4B_4$; we find $a \cong 5.320$ Å and $c \cong 7.448$ Å.

There is evidence of the common impurity phase RhB as well as others ($SmRh_6B_4$, $SmRh_3B_2$, and $SmRhB_4$) in both scans, but in addition, almost every film contains strong impurity peaks at 30° and/or 43° which we have

TABLE I. A compendium of data for a representative selection of sputtered $SmRh_4B_4$ films. The substrate orientation is represented by θ (see text), the film thickness is d, the residual resistance ratio is r_R, and superconducting transition temperature T_c. The rest of the data represent the height of peaks in the θ-2θ x-ray diffraction patterns. Comparisons from sample to sample are not reliable since no attempt was made to calibrate the system each run; however, relative peak heights for each sample are quantitative (keep in mind preferential orientation mentioned in the text). The column labeled "Total" under $SmRh_4B_4$ includes the 101, 110, 102, 200, 112, 201, 211, 103, 212, and 220 reflections. The 202 is *not* included because of the indistinguishable sapphire-substrate 006 reflection for the 0° orientation. The RhB includes the 100, 101, and 102 reflections, the 002 being masked by the usually large RhAl peak at 43°. Impurity phases identified under "Others" include $SmRh_3B_2$ (usually about 90% of "Others"), $SmRh_6B_4$, and the rarely seen $SmRhB_4$. It should be pointed out that there were no consistent trends associated with substrate orientations except the relatively larger 100 RhAl x-ray peak on 90° substrates, which was most evident for concurrent depositions onto substrates of both orientations (not shown in this table).

					$SmRh_4B_4$					RhB	RhAl		Others
Film no.	θ (deg)	d (μm)	r_R	T_c (K)	Total	110	102	211	103		100	110	
2	0	0.4	3.6	1.618	84	22	25	9	9	3	5	≥ 200	20
4	0	0.4	3.2	1.14	52	6	4	15	8	≤ 1	3	≥ 60	3
6	0	0.5	3.8	1.712	136	33	45	15	9	9	25	≥ 400	26
8	0	0.55	3.5	1.759	157	22	23	18	10	17	35	≥ 170	23+30[c]
9	0	0.36	4.2	1.969	140	32	10	20	7	4	13	≥ 270	28
11	0	0.5	4.3	2.135	143	38	3	10	7	3	12	≥ 280	50
27-2	90	0.1	1.7	<0.4	≤ 7	0	≤ 4	0	0	5	8	180	2
30	90	0.96	3.9	1.815	36	5	2	12	≤ 2	20	≤ 1	≥ 45	<1
31-1	90	1.02	4.2	2.112	73	4	5	23	10	7+100[d]	0	≥ 40	11
31-2	90	1.02	5.0	2.332	108	16	5	23	10	6	9	13	7
32-1	90	0.13	2.3	[a]	46	1	1	1	≤ 1	0	12	57	0
33-2	90	1.08	5.1	2.276	16+65[b]	1	1	0	0	3	≥ 50	12	19
38	BN	0.5	-	-	48	≤ 1	4	26	≤ 2	≤ 1	0	0	14
44	90	0.5	4.3	1.899	55	4	10	13	13	4	2	≥ 160	3

[a]The film resistance dropped to 80% of normal value at 0.4 K.

[b]This predominant peak at 41.76° has not been included in the other totals because it is indistinguishable from the [006] sapphire peak for 0° substrate orientation.

[c]Relies on the identification of rarely seen $SmRhB_4$ from one peak at 40.5°.

[d]This [100] RhB peak is anomalously large.

identified as RhAl. The source of this is almost certainly a reaction with the sapphire (Al_2O_3) substrate: after etching away the $SmRh_4B_4$ film, a depression of about 500 Å is found on the substrate, and the RhAl peaks are completely eliminated when BN substrates are used. Unfortunately, ceramic BN substrates are incompatible with many of the desired transport and superconducting measurements, especially tunneling. However, there is strong evidence that the RhAl is confined to the first 1000 Å or so of the film: x rays of 1000-Å-thick films showed none of the right phase, comparable peaks for RhAl, and no superconductivity down to 0.4 K; x rays of 1300-Å-thick films showed very small correct phase peaks and only a partial (20%) transition at 0.4 K. Energy dispersive x-ray measurements on a thick (4000-Å) film show a barely resolved aluminum peak which is at least 10 times smaller than that seen on a thin (1000-Å) film. It is probable that the x-ray intensity for the RhAl is enhanced due to a high degree of preferential orientation; for example, these peaks are occasionally anomalously small, perhaps indicating a slight misorientation of the substrate.

C. Superconducting transition and residual resistance ratio

Most of the samples were cooled in a standard helium-3 cryostat for these measurements. The usual four-probe technique, with very small currents, was used for the measurement of r_R and the resistive transition at T_c. The sample temperature was slowly drifted through the transition to ensure thermal equilibrium.[15] Values of T_c quoted are the midpoints of the resistive transition and are summarized in Table I along with r_R. The transition widths (10%–90%) varied from ~50 to ~200 mK. The resistance ratio at ~77 K correlated well with r_R measured at 4.2 K.

In Fig. 2, T_c is plotted against r_R for our $SmRh_4B_4$ films: these data deviate greatly from the general behavior[18,19] shown for various other superconductors including $ErRh_4B_4$. A discussion of this discrepancy is deferred until a later section.

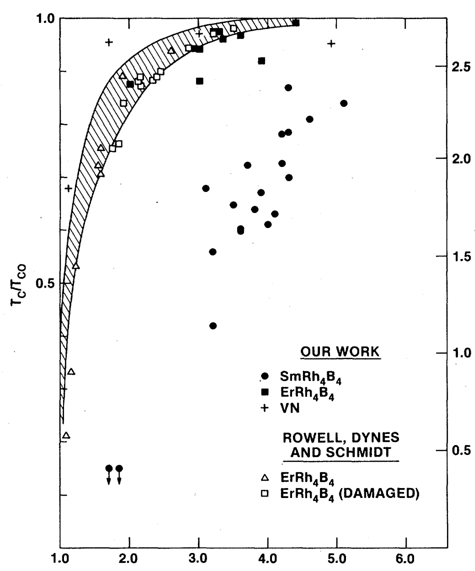

FIG. 2. The variation of T_c with residual resistance ratio r_R for "as made" films of $SmRh_4B_4$. Also shown are $ErRh_4B_4$ films made by us and in Ref. 19 (including radiation-damaged films) and our results for VN films. The universal behavior of $A15$ superconductors is indicated by the shaded region. The scale on the right applies only to the $SmRh_4B_4$ films. T_{c0} is the transition temperature of the bulk material.

III. CRITICAL FIELDS, CURRENTS

The temperature-dependent critical-field curve for bulk $SmRh_4B_4$ shows structure at a temperature T_N associated with the antiferromagnetic ordering.[3] Such measurements for our films yield similar results. On the other hand, measurements of the critical current I_c in zero applied field, for two of the same films, show no recognizable structure at T_N.

For most samples, measurements were made with the field both parallel and perpendicular to the film paying particular attention to the procedures of Ref. 15. In contrast to similarly made $ErRh_4B_4$ films which exhibited large anisotropy,[15] the results on $SmRh_4B_4$ films were very nearly isotropic, with $H_{c\parallel}$ slightly greater than $B_{c\perp}=B_{c2}$. This is consistent with the much smaller[3] magnetic susceptibility of $SmRh_4B_4$. These films exhibit nonhysteretic resistive transitions, indicative of type-II superconductivity, over the entire temperature range, whereas $ErRh_4B_4$ transforms to type I at low temperatures.[15]

In Fig. 3, the perpendicular critical field B_{c2} is shown as a function of temperature for several representative samples with significantly different T_c. In spite of this variation in T_c, the kink structure at ~0.85 K, which is associated with the antiferromagnetic ordering temperature T_N, is relatively constant. Also the magnitude of B_{c2} at low temperatures, $T \lesssim T_N$, is nearly the same for all the film samples and bulk.[3] If one carefully determines the kink position, there is a positive correlation for T_N to increase with T_c (e.g., for $T_c=1.618$ K, $T_N \cong 0.85$ K, and

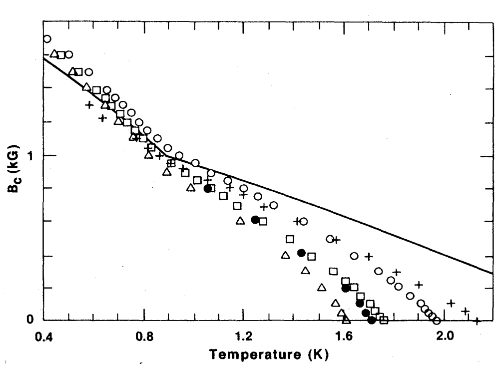

FIG. 3. Critical fields as a function of temperature are plotted for samples (in order of increasing T_c): 2, 6, 8, 9, and 11. Other details of these samples are contained in Table I. Also shown as the solid line is the behavior of the bulk polycrystalline sample of Ref. 3.

for $T_c = 2.332$ K, $T_N \cong 0.95$ K). This correlation was also found between the ferromagnetic ordering temperature T_m and T_c in radiation-damaged $ErRh_4B_4$ films.[20]

IV. IMPURITY PHASES

Generally our films were of high quality when judged by x rays, T_c, and r_R (see Table I). In condensing films of $SmRh_4B_4$, one faces some problems which can be eliminated when arc-melting bulk materials. One would like to have as high a temperature as possible (up to about 1100 °C) for the formation of the correct phase and to achieve a well-ordered material. However, higher temperatures promote the previously mentioned reaction with the sapphire substrate and may result in the preferential loss of the most volatile components of the ternary material. For films made at a significantly lower temperature than the bulk, the problem is further compounded by a reduced diffusion rate and reaction time, and thus we speculate that slight deficiencies or excesses in a particular component can lead to the growth of a nonstoichiometric phase. Some improvements have been noted by intentionally introducing an excess of a particular component (say boron) in the sputtering target.[21] However, it may well be impossible with present technology to produce single-phase films of these ternaries.

A. Effects of impurity phases on our measurements

Before discussing impurity effects on each measurement individually, it should be pointed out that the dominant impurity in the x-ray scans, i.e., RhAl, is expected to behave differently because it is a continuous interfacial layer, presumably about 1000 Å thick at the substrate interface, while other trace impurities may be isolated grains throughout the thickness of the film. Thus the effect of the RhAl should diminish for films of greater total thickness.

The impurity effects on T_c and B_{c2} should be very similar because the measurement of B_{c2} is really a measure of $T_c(B)$. In a sample containing nonsuperconducting impurity phases, the resistive transition occurs when the first $SmRh_4B_4$ superconducting path percolates through the sample length. However, T_c of the superconductivity can be reduced by the proximity effect[22] from neighboring nonsuperconducting impurity phases. The reduction in T_c will be small (large) when the width of the superconducting regions is large (small) compared to the $SmRh_4B_4$ coherence length ξ, which is estimated from B_{c2} to be about 500 Å for the pure material for $T \lesssim 1$ K [see Eq. (2) below]. For the dominant impurity in the x rays, the RhAl interfacial layer, the proximity effect is diminished greatly[22] when the film thickness $d \gg d_N$, ξ where d_N is the thickness of RhAl. This is shown by the results presented in Table I: Films of thickness 0.1 μm (e.g., No. 27-2) were not superconducting down to 0.4 K and the x-ray intensity of the $SmRh_4B_4$ phase was negligible. For a thickness of 0.13 μm (e.g., No. 32-1) the x-ray intensity of the correct phase was relatively larger but the superconducting transition was incomplete (80% of normal resistance) at 0.4 K. On the other hand, *there was no systematic variation in T_c with thickness for films of total thickness between 0.4 and 1.26 μm.* This result implies that proximity with the RhAl interfacial layer has a negligible effect on T_c for film thicknesses $d \geq 0.4$ μm. However, the effect on T_c of other impurity phases, which may be distributed throughout the film, will not necessarily decrease with increasing thickness. The x-ray intensities of these impurities (RhB, $SmRh_3B_2$, $SmRh_6B_4$, and $SmRhB_4$) are generally quite small compared to $SmRh_4B_4$ (see Table I), and the T_c of our films seems most sensitive to the relative total x-ray intensity of the $SmRh_4B_4$ phase, showing little correlation with impurities. However, because of possible preferential orientation the proximity effect cannot be ruled out, and will be discussed in greater detail in the next section.

The effect of the RhAl layer on the measured residual resistance ratio r_R will depend on the value of r_R for the RhAl film (found to be 1.7; see Table I) and the ratio β of the room-temperature phonon resistivity of RhAl to that of $SmRh_4B_4$.[23] Because the measured r_R is always greater than 1.7, the effect of correcting for the RhAl layer is always to *increase* r_R of the $SmRh_4B_4$ film above the measured value. To systematically reverse this requires a RhAl r_R substantially greater than the measured r_R (up to 5), which seems implausible. From resistivity measurements, we estimate $\beta \cong 0.5$, which indicates that the correction to the measured r_R due to RhAl is small but not negligible (as much as 20% for the thinnest samples, i.e., $d \sim 0.4$ μm). However, due to uncertainties in β, and in the thickness and r_R of the RhAl layer for any particular sample, we only quote *measured* values of r_R in Fig. 2 and Table I. Note that such corrections would only *increase* the discrepancy of T_c versus r_R with the universal model (see Fig. 2).

In order to determine the effect of distributed impurity phases on r_R one requires knowledge of the scattering mechanisms restricting the electron mean free path. If scattering due to disorder *within* grains predominates then impurity phases will not affect r_R; however, if grain-boundary scattering by impurity phases predominates, then r_R will be affected. The effect of different types of disorder on the superconducting properties will be discussed in the next section.

B. Effects of impurity phases on tunneling measurements

In the Introduction, we discussed the importance of tunneling measurements to better understand the interaction of superconductivity and magnetism. The effect of the RhAl interfacial layer on tunneling measurements can be greatly reduced by using a total film thickness much greater than ξ. However, impurity phases at the film surface are particularly disastrous for quasiparticle tunneling since the tunnel current is the sum of contributions from the entire junction area. Such measurements are viable only if the impurity phases can be demonstrated to be a small ($\lesssim 1\%$) fraction of the film area.[24] Because of the difficulties of both reducing and measuring impurity phases, quasiparticle tunneling in $SmRh_4B_4$ may have to wait for vacuum tunneling from a sharp point to a single crystal or crystallite of the correct phase.[25] On the other hand, by its very nature, Josephson tunneling of superconducting pairs at zero voltage avoids the complications of normal impurity phases at the surface of the tunnel junction. This is because no supercurrents will flow into these phases.

V. ANOMALOUS T_c VERSUS r_R

Measurements of T_c versus r_R for a large class of superconductors exhibit a universal behavior, first pointed out by Poate *et al.*,[18] and shown as the shaded area in Fig. 2. The samples include "as made" films[15,19] and films progressively damaged by various radiations[19] (α particles, neutrons, etc.). Materials as diverse as $A15$ compounds, $ErRh_4B_4$, and our recent results on VN films exhibit such universal behavior. Although the exact nature of the disorder is unknown, two theoretical concepts have been suggested as being responsible. In the first,[26] 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 model[27] has been suggested, from which the universality of the behavior is more readily understood.

Our measurements for $SmRh_4B_4$ films are also shown in Fig. 2 and they clearly disagree dramatically with the universal behavior discussed above. On the other hand, our $ErRh_4B_4$ films[15] made under nominally identical conditions, follow the universal behavior. Therefore, any explanation of disorder effects must include a reason for the different behavior of these materials. In the following, we shall consider two potential explanations: a proximity effect with impurity phases; and enhanced spin-flip scattering due to disorder. Neither of these are contained in the universal models described above and since they are additional mechanisms of T_c reduction, their presence will not contradict the universal models. It is important to realize that each of our explanations requires a different type of disorder that will be described below. After discussing these individually, an experiment is proposed to determine if the effect is an intrinsic property of $SmRh_4B_4$, like our disorder-enhanced spin-flip scattering model, or the fundamentally less interesting proximity effect.

A. Proximity effect

One possible explanation of Fig. 2 is the presence of impurity phases in the films which could reduce T_c via the proximity effect. It is well known that a superconductor with a long coherence length ξ is more severly affected by proximity to a normal region. It can therefore be understood why the T_c degradation due to the proximity effect could be substantially greater in $SmRh_4B_4$ than in $ErRh_4B_4$: the coherence length estimated from T_c or B_{c2} using standard relations is expected to be about three times larger in $SmRh_4B_4$. Another important consideration for this model to explain Fig. 2 is the necessity to postulate a difference in the grain structure (which gives a stronger proximity effect and greater T_c reduction) as r_R decreases. For example, the reduction of T_c by the proximity effect would be greater for larger normal phase grains, but that is inconsistent with r_R decreasing. On the other hand, if, when r_R decreases, the number of normal phase grains (distributed throughout the film) becomes greater and the size of the superconducting regions between them decreases, then the behavior shown in Fig. 2 could occur as a result of the proximity effect. It should be emphasized, however, that based on this discussion and that of the previous section, it is unlikely that a proximity effect with the RhAl layer causes the variation in Fig. 2.

B. Disorder-enhanced spin-flip scattering

It is well known that spin-flip scattering is much smaller in ordered RRh_4B_4 compounds (R =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 (RE) ions.[28] Such a small overlap must rely on a crystal with a reasonable degree of long-range order. Consequently, any disorder in this perfect lattice structure should increase the overlap, enhance the spin-flip scattering, and lower T_c. Note that only disorder within the $SmRh_4B_4$ grains, and not impurity phases, would contribute to this effect.

As in the case of the above proximity-effect model, it is necessary to demonstrate at least qualitatively that the effect of disorder-enhanced spin-flip scattering is smaller in $ErRh_4B_4$ than in $SmRh_4B_4$. The effect of spin-flip scattering on T_c can be analyzed within the context of the Abrikosov-Gor'kov (AG) theory[29] which predicts a universal behavior for the reduction of T_c with the pair-breaking parameter

$$\alpha=\hbar^{-1}nN(E_F)\mathcal{J}^2(g_J-1)^2J(J+1)\ , \qquad (1)$$

where $N(E_F)$ is the density of states at the Fermi energy E_F, n the concentration of RE atoms, the Landé g factor is g_J, the total angular momentum of the RE ion is J, and $\mathcal{J}$ is the exchange interaction parameter between the conduction electrons and the RE localized moments. By introducing dilute concentrations of various RE ions into $LuRh_4B_4$ (which is nonmagnetic with a superconducting T_c of 11.4 K), MacKay *et al.*[30] measured the relative size of $(dT_c/dn)_{n\to 0}$ for the various RE atoms. For pure $SmRh_4B_4$ and $ErRh_4B_4$ the limit $n\to 0$ is not satisfied, but one can understand qualitatively why T_c is reduced from $LuRh_4B_4$ by about three times more for $SmRh_4B_4$ than for $ErRh_4B_4$, because $(dT_c/dn)_{n\to 0}$ is about four times larger for $SmRh_4B_4$.

The AG theory also forms a basis for understanding the effect of disorder on T_c in these compounds. The goal is to evaluate the change in T_c with disorder; the universal AG curve predicts the dependence of T_c on α, so the dependence of α on disorder is required. Referring to Eq. (1), only $N(E_F)$ and $\mathcal{J}^2$ can be reasonably expected to change with disorder. Severe disorder can affect[26] $N(E_F)$ (see above), but apparently[19,20] only for lower values of r_R than found in our $SmRh_4B_4$ films. However, as pointed out above, the smallness of $\mathcal{J}$, the exchange interaction for overlap of the conduction and localized RE spin electron states, must rely on a reasonable degree of long-range crystalline order. Thus $\mathcal{J}$, and hence α, could change significantly even for mild disorder. To complete such a model would require first-principles calculations in the spirit of Ref. 28 on the effect of disorder on the overlap integral $\mathcal{J}$. However, since the exact nature of the disorder is unclear, we present an *ad hoc* assumption which indicates the plausibility that the *quantitative* differences between $SmRh_4B_4$ and $ErRh_4B_4$ may be reasonably close to the measurements of Fig. 2.

Assuming that the overlap is confined to 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 $\mathcal{J}^2$ will be proportional to $\mathcal{J}^2$. From its definition in Eq. (1), it is therefore clear that the change in α with disorder is proportional to α. Thus 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 $ErRh_4B_4$ and the various $SmRh_4B_4$ samples, T_{c0} is assumed to be the value (11.4 K) for nonmagnetic $LuRh_4B_4$. Thus one finds that $\alpha(dT_c/d\alpha)$ and hence the T_c depression for $SmRh_4B_4$ samples is approximately 5–11 times greater than for $ErRh_4B_4$, where the range covers T_c values for $SmRh_4B_4$ between the bulk value $T_{cb}=2.72$ K and $T_{cb}/2$. This is entirely consistent with the data in Fig. 2.

At first glance, the decrease in the measured T_N with increasing disorder (Sec. III) seems to be contrary to the above model in which $\mathcal{J}^2$ increases with disorder. This is because simple models predict the magnetic ordering temperature T_M to be proportional to $\mathcal{J}^2$. However, inconsistencies in the determination of $\mathcal{J}$ from $(dT_c/dn)_{n\to 0}$ and of T_M for the RRh_4B_4 series led to the suggestion[31] that the lattice crystalline electric field (CEF) has an important influence on magnetic ordering. The large magnetic anisotropies found in many RRh_4B_4 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 $SmRh_4B_4$) due to anisotropy changes and $\mathcal{J}^2$ from the above model. Therefore, disorder enhanced spin-flip scattering cannot be ruled out because T_N decreases with disorder.

C. Proposed radiation-damage experiment

One important difference in the proximity-effect and disorder-enhanced spin-flip scattering models is the nature of the disorder required by the models. As r_R decreases, the proximity-effect model requires the average size of the superconducting regions between normal impurity phases to decrease continuously, whereas the disorder-enhanced spin-flip scattering model requires disorder *within* the superconducting $SmRh_4B_4$ grains to increase. A convenient probe to distinguish these models is radiation damage with protons or α particles, which will only affect disorder within the grains. Thus, the result of damage will be a *weakening* of the proximity effect, since the shorter electron mean free path will lead to a *shorter* coherence length. On the other hand, such damage enhances the spin-flip scattering. Such an experiment could also rule out the possibility of a proximity effect with the RhAl layer.

VI. TEMPERATURE DEPENDENCE OF B_{c2}

The critical fields of our films are shown in Fig. 3, and are qualitatively similar to the bulk measurements (solid line). It has been shown[3] that the bulk $B_{c2}(T)$ can be fit convincingly to the theory of Machida,[8] and we have done the same for our thin-film data. However, we are hesitant to draw conclusions based on these fits: the Machida model[8] has been criticized for violating a spin sum rule,[9] and the use of four parameters[8] permits a great deal of flexibility in fitting data. For example, the critical-field data for $TmRh_4B_4$ (a bell shape, with no kinks) was fitted[32] very well by the Machida model, but required a $\tau_{s.o.}$ (spin-orbit scattering time) approximately 5000 times smaller than that for $SmRh_4B_4$. Therefore, because of the latitude afforded by a four-parameter fit, it is difficult to assess the degree of confidence to be given to the values of these parameters.

A particularly provocative observation is that all samples, including bulk, show very similar behavior below about 1 K even though T_c varies from 1.61 to 2.13 K for films and is 2.72 K for the bulk. Normally (e.g., the BCS theory), the zero-temperature critical field $B_{c2}(0)$ increases with T_c. In the following, we present a simple model showing how $B_{c2}(0)$ may be independent of T_c.

In the Ginzburg-Landau (GL) theory $B_{c2}(0)$ depends on the zero-temperature GL coherence length, $\xi(0)$, according to[33]

$$B_{c2}(0)=\frac{\phi_0}{2\pi\xi^2(0)}\ , \tag{2}$$

where ϕ_0 is the flux quantum. In the dirty limit (i.e., $l \ll \xi_0$ where l is the electron mean free path and ξ_0 the BCS coherence length) it has been shown[33] that $\xi^2(0)\sim\xi_0 l$. Now $\xi_0=hv_F/1.76\pi k_B T_c$ where v_F is the Fermi velocity, so that

$$B_{c2}(0)\cong 0.88\pi k_B T_c/ev_F l\ . \tag{3}$$

Now l^{-1} is proportional to the residual *resistivity* ρ_0 which is just $\rho_{ph}(300\ \mathrm{K})/(r_R-1)$, where ρ_{ph} (300 K) is the phonon scattering contribution to the resistivity at room temperature. Therefore, $B_{c2}(0)\propto T_c/(r_R-1)$. Referring to Fig. 2, one finds T_c has a good approximation to a linear dependence on r_R-1 so, the constant $B_{c2}(0)$ is not surprising.

VII. SUMMARY OF RESULTS AND CONCLUSIONS

In this section, the results and conclusions are summarized in four categories: film quality, anomalous T_c versus r_R, critical fields, and tunneling measurements.

A. Film quality

Triode sputtering has been used to produce high-quality films of $SmRh_4B_4$ as judged by r_R, x rays, and T_c. There is no demonstrated correlation of the superconducting properties with trace impurity phases found in the x rays, nor the layer of RhAl found at the substrate interface.

However, a proximity effect with impurity phases distributed throughout the film cannot be ruled out.

B. Anomalous T_c versus r_R

Some films have lower, but still respectable, values of r_R (as low as 3.2) and these exhibit lower T_c. The variation of T_c with r_R is significantly different from the universal behavior of $A15$ and $ErRh_4B_4$ superconductors. Two possible models are presented which could explain this: a fundamentally less interesting proximity effect with distributed impurity phases; and a new effect—disorder-enhanced spin-flip scattering. A radiation-damage experiment is proposed which can distinguish between the proximity effect and an intrinsic property of $SmRh_4B_4$, since the nature of the disorder required is different.

C. Critical fields

Measurements of $B_{c2}(T)$ on various films are similar to bulk $SmRh_4B_4$, but have lower T_c in zero field. At low temperatures, B_{c2} is independent of T_c and a simple model is presented to explain this.

D. Tunneling

In the Introduction, it was shown that tunneling could provide a very important tool to distinguish between various theories of B_{c2}, by looking directly at the superconducting condensation energy E_c in zero field. Uncertainties in the amount of distributed impurity phases at the junction barrier will make quasiparticle (finite voltage) tunneling difficult to interpret. However, Josephson tunneling at zero voltage avoids the nonsuperconducting impurity phases and is thus recommended for such a study.

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