Observation of Enhancement of the Superconducting Order Parameter in the Coexistent Antiferromagnetic State of $SmRh_4B_4$

Ruggero Vaglio,[a] B. D. Terris, J. F. Zasadzinski,[b] and K. E. Gray
Materials Science and Technology Division, Argonne National Laboratory, Argonne, Illinois 60439
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The first measurements of Josephson tunneling into both the paramagnetic and antiferromagnetic phases of $SmRh_4B_4$ are reported. The results show that the superconducting order parameter of $SmRh_4B_4$ is enhanced by the antiferromagnetic order occurring below the Neél temperature of about 0.87 K. On the basis of this observation, the applicability of various proposed theories of coexistent superconductivity and antiferromagnetism can be determined for $SmRh_4B_4$.

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The microscopic coexistence of superconducting and antiferromagnetic order is displayed in several ternary compounds.[1] The effect of the competing order parameters is manifested in the unusual temperature dependences of the critical magnetic field, $B_{c2}(T)$, compared to that of an ordinary (e.g., BCS) superconductor. Theoretical models have been presented[2–7] that consider various mechanisms through which the antiferromagnetism affects the superconductivity and that can explain the various temperature dependences of B_{c2}. However, the superconducting order parameter, ψ, is a more direct measure of the effect of antiferromagnetism on superconductivity and can be used to distinguish between the various proposed models.

We report the first measurements of Josephson tunneling into both the paramagnetic and the antiferromagnetic phases of $SmRh_4B_4$ thin films. It is well known that the Josephson current is sensitive to the superconducting order parameters in each electrode,[8] and our measurements show that ψ of $SmRh_4B_4$ is enhanced by the antiferromagnetic order occurring below the Neél temperature, $T_N \sim 0.87$ K.

The films of $SmRh_4B_4$ were made by dc sputtering using a Plasmax triode sputtering source. Details of film preparation and characterization along with measurements of $B_{c2}(T)$ can be found in the work of Zasadzinski *et al.*[9] The $B_{c2}(T)$ data show a stronger inverse temperature dependence below $T_N \approx 0.87$ K and agree qualitatively with bulk measurements.[10] Films 0.4 to 1.0 μm thick were deposited onto single-crystal sapphire substrates at temperatures from 800 °C to 1000 °C. After allowing the substrate to cool to ~ 250 °C (~ 25 min), a thin layer (30–40 Å) of Lu was sputtered from a second gun without breaking vacuum. Once the substrate reached ~ 50 °C ($\sim 2\frac{1}{2}$ h), the Lu was oxidized in ~ 250 mTorr of pure oxygen for ~ 10 min to form the tunnel barrier.[11] The vacuum chamber was then opened, the sample removed, and its edges were coated with collodion. Three Pb counterelectrodes (~ 5000 Å thick) were electron-beam deposited onto the substrate thus forming three tunnel junctions (areas were ~ 0.7 mm^2).

The junctions were studied in a conventional ^{3}He cryostat. The current-voltage characteristics $I(V)$ showed gap structure of the Pb including its well-defined phonon structure, but with significant leakage ($\sim 50\%$). There was no evidence for a peak in the density of states of the $SmRh_4B_4$, but this may be due to gaplessness because of the relatively large magnetic pair breaking; i.e., T_c is significantly reduced from the nonmagnetic $LuRh_4B_4$ value of 11.4 K. For this reason we report measurements of the zero-voltage Josephson current, I_c, which has two important advantages: It is insensitive to nonsuperconducting impurity phases in the sputtered film; and it is directly related to the order parameter of the $SmRh_4B_4$ phase.

The maximum Josephson current was measured as a function of applied parallel magnetic field at several temperatures below T_c of $SmRh_4B_4$ (Fig. 1). These diffraction patterns are not ideal, but exhibit a well-defined central maximum with several secondary peaks of reasonably appropriate heights. The deviations from ideal behavior can be attributed to any or all of the following: a nonuniform junction width perpendicular to B; a nonuniform tunneling barrier; or trace secondary phases in the $SmRh_4B_4$ film. No additional structure was seen in the diffraction pattern on passing through T_N.

Figure 2 shows the maximum I_c of the central peak as a function of temperature for one of our junctions. Other junctions showed quantitatively similar I_c enhancement below T_N (inset of Fig. 2), but the general T dependence was slightly concave upward rather than the strict linear dependence shown in Fig. 2.

The order parameter of the Pb counterelectrode

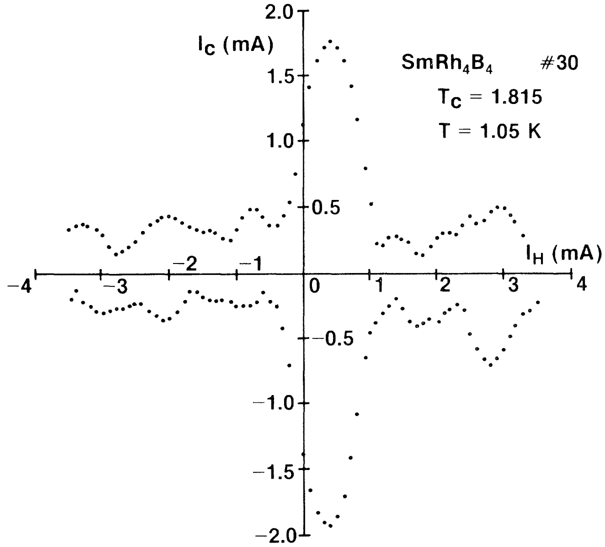

FIG. 1. Typical Josephson diffraction pattern obtained by measuring I_c as a function of a small parallel field (measured by the magnet current I_H, where the magnet constant is ~ 100 G/A).

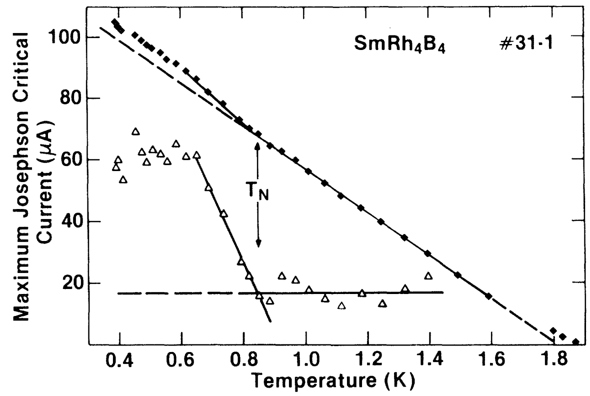

FIG. 2. The maximum Josephson current vs temperature. Inset: Difference of data from straight line extrapolated from above T_N (scale $\times 10$).

is approximately constant in this temperature range, so that the small but distinct increase in $I_c(T < T_N)$ over the value extrapolated from above T_N shows that the strength of the superconductivity and hence ψ in $SmRh_4B_4$ is enhanced for $T \leq T_N$. That the temperature dependence of $I_c(T)$ follows ψ closely will be shown below for several simple models (see Fig. 3). Thus for $T < T_N$, our I_c data indicate that ψ is enhanced by $\sim 5\%$, an amount consistent with the decreased pair breaking near T_N estimated by Ramakrishnan and Varma.[3]

Following Ramakrishnan and Varma, a qualitative explanation for decreased pair breaking near and just below T_N can be given. In the quasistatic scattering limit,[3] the dimensionless pair-breaking parameter is

$$\rho = (3\mathscr{J}^2/\pi) \sum_{\vec{q}} \phi(\vec{q}) \chi(\vec{q}), \tag{1}$$

where $\mathscr{J}$ is the exchange interaction parameter between conduction electrons and the localized (Sm) moments, $\chi(\vec{q})$ is the spin susceptibility of those localized moments, and

$$\phi(\vec{q}) = \frac{1}{N(E_F)V^2} \sum_{\vec{k}} \delta(\epsilon_{\vec{k}} - E_F)\,\delta(\epsilon_{\vec{k}} - \epsilon_{\vec{k}+\vec{q}}) \tag{2}$$

is the joint density of states for the conduction electrons, which is a measure of the coupling strength of the electrons to the various wave-vector components of $\chi(\vec{q})$. Here $N(E_F)$ is the density of electron states at the Fermi energy E_F and V is the volume. Note that $\chi(\vec{q})$ satisfies[3] a sum rule for

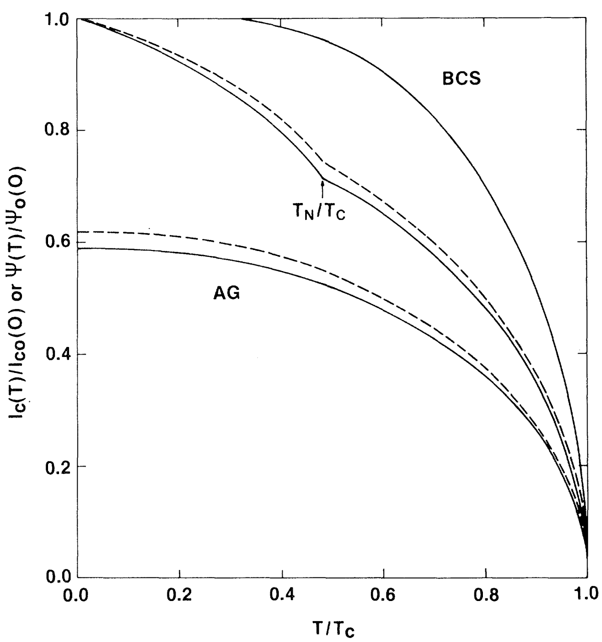

FIG. 3. Order parameter ψ (solid curves) and maximum Josephson current I_c (dashed curves) for three models of superconductors (top to bottom): BCS; modified AG as explained in text; AG. For I_c, the counterelectrode is assumed to have a much higher T_c, so that its order parameter is constant over the temperature range shown. The AG curve and the modified AG curve are normalized by the same values of $I_{c0}(0)$ and $\psi_0(0)$. For the BCS case, ψ and I_c are indistinguishable on this plot.

$T > T_N$:

$$N^{-1} \sum_{\vec{q}} \chi(\vec{q}) = nS(S+1)/3k_B T, \qquad (3)$$

where N is the total number of localized moments of spin S and $n = N/V$. For rare-earth ions (like Sm) in which the orbital angular momentum L is not quenched, S should be replaced by its projection on the total angular momentum vector $\vec{J} = \vec{L} + \vec{S}$ of the Hund's rule ground state. This will not affect our qualitative arguments, and is ignored here for simplicity.

For $T \gg T_N$, the spins are essentially independent, and in the quasistatic limit, $\chi(\vec{q})$ equals[3] $nS(S+1)/3k_B T$, independent of $\vec{q}$. Further, since it can be shown that $\sum_{\vec{q}} \phi(\vec{q}) = N(E_F)$, Eq. (1) reduces to the Abrikosov-Gor'kov (AG) expression[12]:

$$\rho_{AG} = \mathscr{J}^2 N(E_F) nS(S+1)/\pi k_B T. \qquad (4)$$

As T approaches T_N there is a shift in the spectral weight of $\chi(\vec{q})$ towards $\vec{q} = \vec{G}$, where $\vec{G}$ is the wave vector of the ordered antiferromagnetic state. This results in emphasizing the $\vec{q} = \vec{G}$ component of $\phi(\vec{q})$ at the expense of others. The change in ρ from ρ_{AG} (expected to be small[3]) therefore depends on the details of $\phi(\vec{q})$, which in turn depends on the Fermi surface (FS).

For a spherical FS, $\phi(\vec{q})$ is maximum at $\vec{q} = 0$ and decreases as q^{-1} near $q = k_F$, the Fermi momentum. In such a case, the shift in spectral weight of $\chi(\vec{q})$ towards $\vec{q} = \vec{G}$ would be to a region of lower coupling strength, $\phi(\vec{G})$, and ρ would decrease. However, if there is nesting of the FS near $\vec{q} = \vec{G}$, then $\phi(\vec{q})$ will peak there and pair breaking can increase as $T \rightarrow T_N$. A detailed treatment of pair breaking when $\phi(\vec{q})$ peaks near $\vec{G}$ is given in Refs. 5 and 6.

Our $I_c(T)$ data indicate that ψ is enhanced below T_N so that pair breaking is decreased. This implies that nesting of the FS does not dominate the effect of antiferromagnetic order on $SmRh_4B_4$, although it may do so in other antiferromagnetic superconductors.[1] It should be noted that the abrupt (within about 0.1 K of T_N) enhancement in $I_c(T)$ should result from elastic or quasistatic processes only. The shift of spectral weight of $\chi(\vec{q})$ from elastic to inelastic scattering due to spin waves below T_N can also lead to decreased pair breaking,[3] but only for T far below T_N.

Theoretical models which treat only electromagnetic interactions in describing the $B_{c2}(T)$ of antiferromagnetic superconductors[4] have neglected the effect of pair breaking on the superconducting condensation energy (or ψ). Our results indicate that such an assumption is invalid for $SmRh_4B_4$.

In addition to establishing the enhancement of ψ below T_N, it would be nice to use the $I_c(T)$ data for a more definitive test of the theories. In order to do so, it is necessary to relate ψ to I_c. Baratoff[13] has shown how to calculate I_c from ψ; however, it is not clear whether the inverse process is possible. Thus we calculate $I_c(T)$ by Baratoff's method for three models of $\psi(T)$: the BCS model, the AG model,[12] and an approximation to the previously discussed model of Ramakrishnan and Varma for antiferromagnetic superconductors[3] in which nonzero q values are ignored.[2] In Fig. 3 the results for $I_c(T)$ and $\psi(T)$ are plotted for these models and clearly in all cases the temperature dependences of I_c and ψ are nearly identical.

The middle curve shown in Fig. 3 was derived with the assumption that $\phi(\vec{q})$ is a sharply peaked function near $\vec{q} = 0$, so that

$$\rho = 3\mathscr{J}^2 N(E_F) \chi_0/\pi, \qquad (5)$$

where χ_0 is the magnetic susceptibility measured in a magnetization experiment on the antiferromagnet. This model[2] assumes a mean-field Curie-Weiss behavior for χ_0, and qualitatively reproduces[10] the enhanced ψ and B_{c2} below T_N. However, the model has been criticized[3] for ignoring finite-q contributions. The results shown in the middle curve of Fig. 3 use parameters arrived at by independently fitting[10] $B_{c2}(T)$, and seem to overestimate ($\sim \times 2$) the experimental enhancement of I_c and hence ψ (but see the discussion below on the proximity effect).

Because of the close relationship between $I_c(T)$ and $\psi(T)$ shown in Fig. 3, it is tempting to interpret the I_c data of Fig. 2 as being directly proportional to ψ. However, a general property of second-order phase transitions is that $(d\psi/dT)_{T_c} \rightarrow \infty$, as in the examples shown in Fig. 3. Thus the finite $(dI_c/dT)_{T_c}$ shown in Fig. 2 is inconsistent with $I_c \propto \psi$. One possible cause of this finite slope is inhomogeneities[14] in T_c; however, it seems unlikely that a reasonable amount of smearing will produce a constant $d\psi/dT$ over the wide temperature range of Fig. 2, starting with curves such as those in Fig. 3. Perhaps a more acceptable explanation can be found in the proximity effect, caused for example by any unoxidized part of the thin layer of Lu used for the artificial tunnel barrier.

Several theoretical approaches have been used to analyze $I_c(T)$ for such proximity junctions. Recently Gallagher[15] has outlined a complete microscopic theory which approximately accounts for mean-free-path effects. Previously, Gilabert *et al.*[16] had used the more restrictive McMillan tunneling model[17] of the proximity effect to analyze their results. Earlier experiments were analyzed[18] within the framework of the Ginzburg-Landau theory which is valid near T_c and in the dirty limit. However, the effect of *all of these* is to transform the concave-downwards curves of $I_c(T)$ shown in Fig. 3 into linear behavior over a wide temperature range (as shown in Fig. 2), or into concave-upwards curves (as found by us for other junctions of $SmRh_4B_4$) depending on the parameters which measure the importance of the proximity effect.

The Ginzburg-Landau approach has the simplification that only the order parameter of $SmRh_4B_4$ is required rather than the Green's function. Since the dirty limit is satisfied in our films, the results near T_c should be valid. Direct calculation using the method of Ref. 18, and the middle curve of Fig. 3 for the order parameter, shows quite acceptable agreement with the data in Fig. 2 between T_N and T_c. Hopefully, further work can reduce or eliminate the proximity effect by making better barriers.

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[a]Permanent address: Dipartimento di Fisica, Università di Salerno, Salerno, Italy.

[b]Permanent address: Department of Physics, Illinois Institute of Technology, Chicago, Ill. 60616.

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