# Investigation of the Magnitude and Range of the Ruderman-Kittel Interaction in $SmRh_4B_4$ and $ErRh_4B_4$

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The superconductive and magnetic transition temperatures taken together are shown to provide a unique probe which separately determines *both* the magnitude and range of the Ruderman-Kittel interaction in the $RRh_4B_4$ magnetic superconductors ($R = \mathrm{Er,Sm}$). Experimentally, an unexpected peak is found in the antiferromagnetic-ordering temperature of $SmRh_4B_4$ versus electron mean free path, while for $ErRh_4B_4$ the ferromagnetic-ordering temperature decreases monotonically. These qualitative features, as well as the quantitative differences between $SmRh_4B_4$ and $ErRh_4B_4$, are in excellent agreement with calculations using a mean-free-path–dependent Ruderman-Kittel interaction.

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We present a novel method for probing the details of the Ruderman-Kittel-Kasuya-Yosida interaction (RKKY).[1] By studying the effect of disorder on the magnetic superconductors $ErRh_4B_4$ and $SmRh_4B_4$, we have investigated the behavior of both the magnitude and the range of the RKKY interaction. The *magnitude* of the RKKY interaction is probed by the on-site spin-flip scattering which affects the superconducting transition temperature, $T_c$, through pair breaking, while the magnetic-ordering temperature depends on the *range* of the oscillatory RKKY interaction as well as on its (on-site) magnitude. It was previously proposed that disorder reduces $T_c$ because of disorder-enhanced spin-flip scattering.[2] The exponential damping of the range of the RKKY interaction due to finite mean free path, $l$, is well documented in the literature.[3] Hence, by measuring the disorder dependence of *both* the superconducting and magnetic transition temperatures, we are able to probe separately the two aspects of the RKKY interaction, and thus explain the detailed behavior of the magnetic-ordering temperature, $T_M$, in $ErRh_4B_4$ and the Néel temperature, $T_N$, in antiferromagnetic $SmRh_4B_4$. In our calculations, we have kept the Fermi momentum $k_F$ and the dependence of $l$ on resistivity the same for each material, using values which are very close to previous estimates and free-electron values.

Both $T_c$ and $T_N$ ($T_M$) for the antiferromagnetic superconductor, $SmRh_4B_4$,[2] and the ferromagnetic superconductor, $ErRh_4B_4$,[4] have been studied previously as a function of radiation damage (which induces disorder and shortens $l$). In each material $T_c$ was found to decrease as a function of radiation dose, though much faster in $SmRh_4B_4$. A model[2] has been suggested which explains this as an enhancement of the *on-site* exchange interaction $\mathcal{J}$ through a new disorder effect on superconductivity due to the presence of magnetic moments. However, the magnetic transition temperature was found to decrease with dose in $ErRh_4B_4$, and to peak with dose in $SmRh_4B_4$, whereas the disorder-enhanced $\mathcal{J}$ model would predict only an increase.

In addition to increasing the magnitude of the RKKY interaction, via the enhancement of $\mathcal{J}$, disorder will also reduce the range of the RKKY interaction. de Gennes[5] proposed that for finite $l$, the RKKY interaction, which is given by the product of the on-site interaction $\mathcal{J}$ and an oscillatory term, will also be damped by $\exp(-r/l)$. In a mean-field approach, the Curie-Weiss temperature ($T_M$ for ferromagnetic and $-T_N$ for antiferromagnetic systems) is given by[6]

$$T_M = -(k_F^3/3\pi)N(E_F)\mathcal{J}^2(g_J-1)^2J(J+1)\sum_r F(2k_F r)\exp(-r/l)s(\mathbf{q}\cdot\mathbf{r}),$$

where

$$F(2k_F r) = \frac{2k_F r\cos(2k_F r) - \sin(2k_F r)}{(2k_F r)^4},$$

$N(E_F)$ is the density of states at the Fermi surface, $g_J$ the Landé $g$ factor, $J$ the total angular momentum, and the sum is over the magnetic-ion lattice sites. The quantity $s(\mathbf{q}\cdot\mathbf{r})$ gives the sign of the spin direction at the position $\mathbf{r}$ for a presumed magnetic ordering described by the vector $\mathbf{q}$. We have taken

$$s(\mathbf{q}\cdot\mathbf{r}) = \cos(\pi x q_x/a)\cos(\pi y q_y/a)\cos(\pi z q_z/c).$$

Thus $\mathbf{q} = (0,0,0)$ corresponds to ferromagnetism, and other $\mathbf{q}$ values to various types of antiferromagnetism. For antiferromagnetic $SmRh_4B_4$, the results are relatively insensitive to the choice of $\mathbf{q}$, as discussed below. To test this model, $l$ was determined for various samples from the residual resistivity with use of the free-electron model[7] for $ErRh_4B_4$ (films[4] and bulk[8]), and for $SmRh_4B_4$ bulk.[9] For the $SmRh_4B_4$ films,[2] $l$ was determined from the resistivity ratios ($r_R$), under the assumption that $SmRh_4B_4$ and $ErRh_4B_4$ films with the same $r_R$ would have the same

 

$l$. After computation of the RKKY sum, it was found that the best fit was achieved by the universal increase of $l$ to $\sim 12\%$ above its free-electron value.

Plots of $T_c$ and $T_N$ versus disorder (which is represented by $1/l$) are shown in Fig. 1 for a bulk sample[9] and radiation-damaged thin films of $SmRh_4B_4$. Also included are $T_N$ for each sample as calculated (pluses) by the procedure outlined below. Experimentally, $T_N$ comes from the inflection point in the superconducting critical field curve (see Ref. 2). Since previous work showed that $\mathcal{J}$ increases with disorder, *part* of the initial increase of $T_N$ vs $1/l$ can be attributed to changes in the on-site interaction $\mathcal{J}$ through Eq. (1). The value of $\mathcal{J}$ for each sample is chosen to achieve the experimentally determined reduction of $T_c$ from $T_{c0}$ by use of the Abrikosov and Gorkov theory with crystalline electric fields (CEF) included,[10] where $T_{c0}=8.95$ K is the expected value for "nonmagnetic" $SmRh_4B_4$.[11] (The overall conclusions, which depend only on the *relative changes* in $\mathcal{J}$ with disorder, would be unchanged if we were to omit CEF or use the $T_{c0}=11.4$ K of $LuRh_4B_4$. Small changes in $k_F$, as discussed below, can easily compensate for changes due to different choices of CEF or $T_{c0}$.) To account for $\mathcal{J}$ and the de Gennes factors, $T_N$ and $T_M$ are normalized by $(g_J-1)^2J(J+1)\mathcal{J}^2$ and plotted in Fig. 2 (with use of $T_{c0}=9.9$ K for $ErRh_4B_4$). The presence (absence) of a peak in $T_N$ for $SmRh_4B_4$ ($T_M$ for $ErRh_4B_4$) can be understood from the oscillatory nature of the RKKY interaction, with use of a simple shell model. Assume the nearest- and next-nearest-neighbor contributions to the sum are antiferromagnetic and ferromagnetic, respectively, for $SmRh_4B_4$. Then since the *next*-nearest-neighbor contribution dies out quicker as a result of $\exp(-r/l)$, the *net* antiferromagnetic interaction can be initially enhanced. For larger $1/l$, the nearest-neighbor antiferromagnetic contribution is decreased sufficiently such that $T_N$ decreases. For the ferromagnetic $ErRh_4B_4$, both nearest- and next-nearest-neighbor contributions are ferromagnetic, in this model, since nearest-neighbor contributions are reversed because the spins are all parallel. Therefore, $T_M$ is expected to decrease and this is shown in Fig. 2.

A number of comments should be noted concerning the above calculations:

(i) The only adjustable parameter is $k_F$, and the values of $k_F$ were obtained by first choosing $k_F$ of $SmRh_4B_4$ to fit the data in the clean limit, and then adjusting $k_F$ of $ErRh_4B_4$ so that the relative magnitudes of the calculated curves match experiment. The value found for both $SmRh_4B_4$ and $ErRh_4B_4$ is 1.568 $Å^{-1}$, which is in close agreement with previous estimates[12]

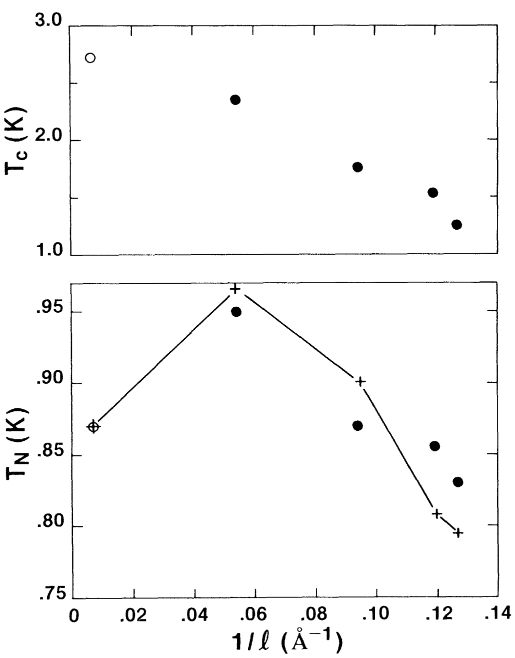


FIG. 1. The superconducting ($T_c$) and antiferromagnetic ($T_N$) ordering temperatures for bulk $SmRh_4B_4$ (open circles) and films (filled circles) vs inverse mean free path ($1/l$). The pluses are calculated values of $T_N$, scaled to match the experimental bulk value.

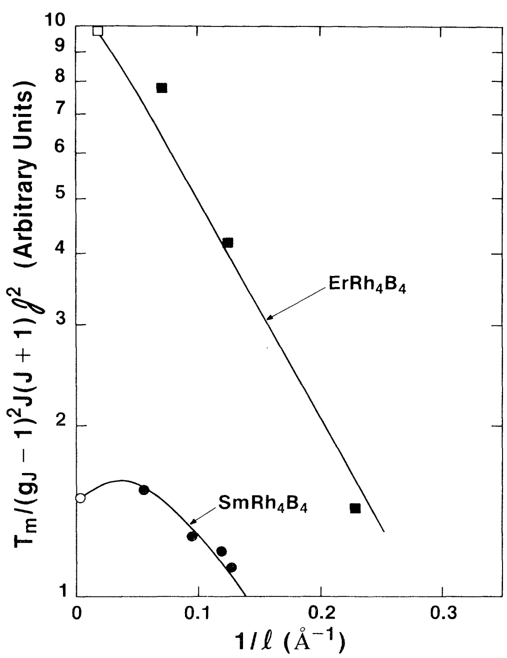


FIG. 2. The magnetic ordering temperatures ($T_M$ for $ErRh_4B_4$, $T_N$ for $SmRh_4B_4$) divided by the de Gennes factor and the exchange interaction $\mathcal{J}^2$ vs inverse mean free path ($1/l$). The open circles are for bulk samples (Refs. 8 and 9); the filled circles for films (Refs. 2 and 4). The calculated values are shown by the solid lines, as described in the text, and scaled at *one* point, the bulk value of $SmRh_4B_4$.

of 1.6 $Å^{-1}$. For $SmRh_4B_4$, a small increase ($\sim 0.001$ $Å^{-1}$) in $k_F$ produces a large increase in the slope ($\times 2$) of the downward turn for small $1/l$. Small changes in $k_F$ also produce overall vertical shifts in both curves, but do not affect the slopes in the dirty limit. An increase in $k_F$ of $ErRh_4B_4$ of $\sim 1\%$ shifts the curve downward by $\sim 30\%$. Therefore, while calculation of exact values for the magnetic transition temperatures would be sensitive to $k_F$, our description of the general features is not. Also, since many approximations are used, the absolute precision of $k_F$ is artificial and the absolute values of $T_M$ and $T_N$ from Eq. (1) are not reliable.

(ii) The lattice sum requires the magnetic ordering vector **q**. For ferromagnetic $ErRh_4B_4$ clearly $\mathbf{q} = (0,0,0)$, while for antiferromagnetic $SmRh_4B_4$, the **q** is unknown. We have assumed it to be (1,1,1), but other choices of **q**, such as (0,0,1), yield similar results. The signs of the calculated sums were found to be consistent with the chosen **q**, i.e., the sum for $ErRh_4B_4$ predicted ferromagnetism and that for $SmRh_4B_4$, antiferromagnetism.

(iii) In the above analysis, modifications to the RKKY interaction based on superconductivity[13] have been ignored. This is strictly valid for $SmRh_4B_4$ since $T_N$ was measured at the normal-superconducting (second-order) phase transition at $H_{c2}$, i.e., in the *normal state*. Tunneling measurements[14] on such films reveal essentially the same Néel temperature in the superconducting state indicating that modifications[13] to the RKKY interaction, at least in this case, are small. This is perhaps due to strong spin-flip and/or spin-orbit scattering reducing the effect of superconductivity on the RKKY interaction.[15] For $ErRh_4B_4$ the experimental situation is complicated by the first-order transition at $T_M$. In addition, the difference in RKKY interaction between the normal and superconducting states may be expected to be greater than in $SmRh_4B_4$, since the spin-flip scattering is weaker in $ErRh_4B_4$, and since superconductivity is expected to have a greater effect on ferromagnetism.[13] This general contention is supported by lattice-sum calculations for this system, using the RKKY interaction as modified by the presence of superconductivity.[16] However, normal-state magnetization measurements on $ErRh_4B_4$[17] suggest that the effect of superconductivity on $T_M$ is not large in this case either.

(iv) Crystalline electric fields are known to play a role in determining the magnetic properties of these materials,[18] resulting in magnetic anisotropies. This lowering of magnetic dimensionality can lead to an enhancement of the magnetic transition temperature.[19] Hence, changes in the CEF due to disorder may play some role in the variation of $T_M$ or $T_N$. This has not been included, since there is no clear way to model the effects of disorder on the CEF.

(v) The exact nature of the radiation-induced disorder is unknown. The behavior of $T_c$ vs $r_R$ is the same in the as-made and radiation-damaged films, and x rays of the damaged films show no change in the amount of trace impurity phases as a function of dose. This suggests that the disorder is in the $SmRh_4B_4$ phase grains.

In conclusion, we have investigated the details of the RKKY interaction by looking at both the superconducting and the magnetic transition temperatures of $ErRh_4B_4$ and $SmRh_4B_4$. We have found a novel method for separately studying the magnitude and range of the RKKY interaction. The magnitude $\mathcal{J}$ is probed by the behavior of $T_c$ while the range is probed by $T_M$. A peak in $T_N$ vs $l$ is found for $SmRh_4B_4$ and is shown to result from the competition between nearest-neighbor (antiferromagnetic) and next-nearest-neighbor (ferromagnetic) interactions.

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