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COEXISTENCE OF SUPERCONDUCTIVITY AND ANTIFERROMAGNETIC ORDER IN $SmRh_4B_4$

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The ternary rare earth compound $SmRh_4B_4$ has been studied by means of upper critical field, low temperature specific heat, and static magnetic susceptibility measurements. A lambda-type specific heat anomaly, a discontinuity in the slope of the upper critical field versus temperature curve, and a cusp-like feature in the magnetic susceptibility suggest that superconductivity and long-range antiferromagnetic order coexist in $SmRh_4B_4$ below 0.87 K. Machida's theory for antiferromagnetic superconductors provides a good description of the upper critical field data, while the magnetic susceptibility data can be represented as the sum of a Curie-Weiss term with $\mu_{eff} = 0.632\,\mu_B$ and $\theta_p = -1.93$ K and a temperature independent Van Vleck contribution.

The class of ternary rare earth (RE) compounds $RERh_4B_4$ has recently been investigated in order to study the interaction between superconductivity and long-range magnetic order. The compounds are ferromagnetic for RE = Gd, Tb, Dy and Ho and superconducting for RE = Sm, Nd, Er, Tm, and Lu.[1,2] Furthermore, $ErRh_4B_4$ exhibits re-entrant superconductivity, wherein the superconductivity is destroyed by the onset of long-range ferromagnetic ordering of the Er^{3+} magnetic moments at a temperature T_{c2} below the superconducting transition temperature T_{c1}.[3,4] Recently we reported the results of an investigation of $NdRh_4B_4$ which revealed two lambda-type anomalies in the heat capacity data, indicative of two phase transitions below the superconducting transition temperature.[5] Features in the upper critical field vs. temperature curve and the static magnetic susceptibility data suggest that the phase transitions are magnetic, and therefore that superconductivity and long-range magnetic order coexist in this compound. However, the presence of impurity phases prevented an unambiguous determination of the exact nature of the magnetic ordering.

The observation of superconductivity and magnetism in both $ErRh_4B_4$ and $NdRh_4B_4$ suggests that the other superconducting $RERh_4B_4$ compounds containing localized magnetic moments might also order magnetically. In this communication we report upper critical field, specific heat, and static magnetic susceptibility data for $SmRh_4B_4$. The results indicate that long-range antiferromagnetic order and superconductivity coexist in this material below 0.87 K.

Two samples of $SmRh_4B_4$ were synthesized by arc melting the high purity elements under argon. The samples were annealed at 1200°C for more than a week, followed by an additional week at 800°C. The first sample was used for four-probe ac electrical resistance and static magnetic susceptibility measurements. In the former experiment a long parallelepiped-shaped sample aligned parallel to various applied magnetic fields was cooled using a He^3-He^4 dilution refrigerator to obtain temperatures from below 0.07 to 10 K. The magnetic susceptibility data were taken between 0.7 and 294 K using a Faraday magnetometer. Heat capacity data for the second sample were obtained between 0.5 and 36 K with a He^3 semi-adiabatic calorimeter using a standard heat-pulse technique.

Figure 1 shows the electrical resistance

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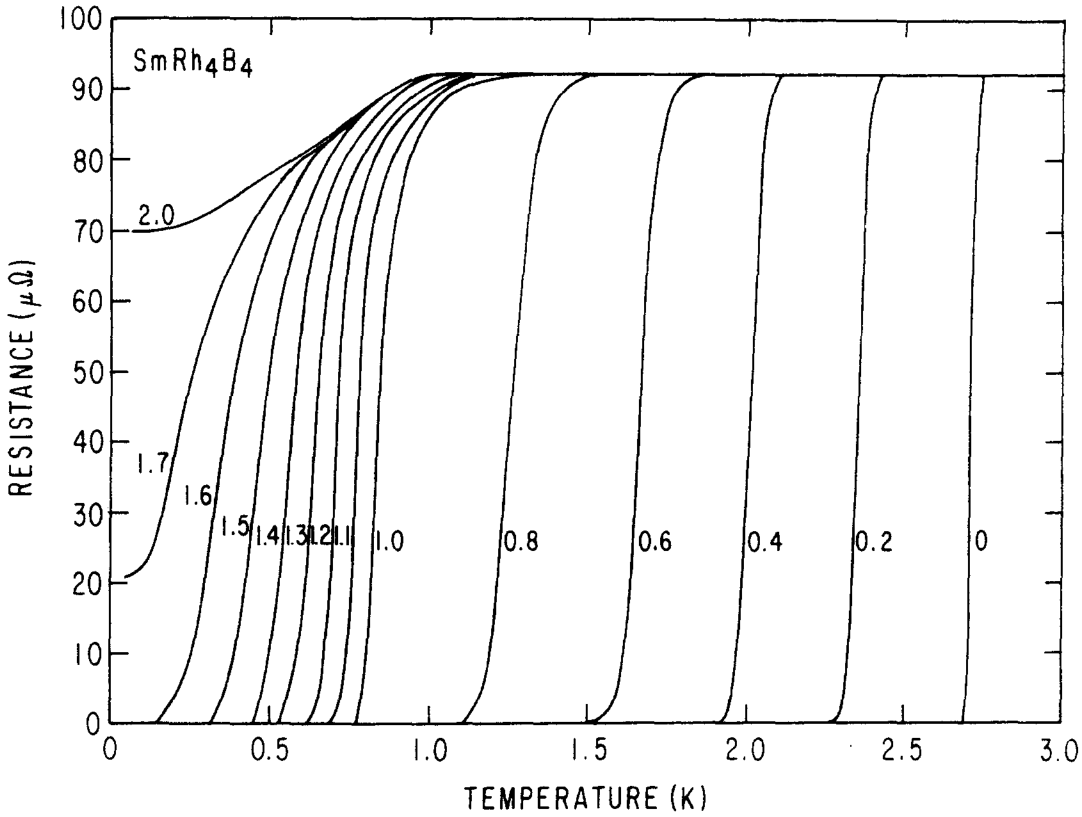

Fig. 1 ac electrical resistance versus temperature for $SmRh_4B_4$ in applied magnetic fields between 0 and 2 kOe.

vs temperature in various applied magnetic fields for $SmRh_4B_4$. In all fields measured below its zero-temperature upper critical field $H_{c2}(0) \sim 1.85$ kOe, the sample exhibits only a single normal to superconducting state transition. The normal state resistance, however, markedly decreases below 0.9 K, indicative of some type of phase transition below the zero field superconducting transition temperature $T_c = 2.72$ K. The upper critical field H_{c2} vs temperature data shown in Fig. 2 seem to support this hypothesis. The transition temperatures were defined as the temperatures at which the sample resistance became fifty percent of the normal state value. The H_{c2} vs temperature curve shows a sharp discontinuity in its slope at approximately 0.85 K; below this temperature H_{c2} is considerably larger than would be expected from the high temperature portion of the curve.

Shown in Fig. 3 is the heat capacity C vs temperature data for $SmRh_4B_4$ between 0.5 and 9 K in zero applied magnetic field. The data reveal a small jump in the heat capacity at the superconducting transition temperature (T_c = 2.72 K) and a pronounced lambda-type anomaly which peaks at a temperature $T_\lambda \cong 0.87$ K. Comparison of the heat capacity data with the upper critical field data shows that T_λ corresponds closely with the temperature at which the discontinuity in the slope of the H_{c2} vs temperature curve occurs. Subtracting the electronic and lattice contributions to the specific heat of the isostructural nonmagnetic compound $LuRh_4B_4$ results in a magnetic entropy between 0 and 16 K of $S_{mag} \simeq R \ln 2$, which suggests that the crystal field ground state of the Sm^{3+} ions is a doublet.

A plot of the reciprocal molar magnetic susceptibility χ_M^{-1} vs temperature for $SmRh_4B_4$ is shown in Fig. 4. The data cannot be described by a simple Curie-Weiss law, which is consistent with the Sm^{3+} ions having relatively low-energy angular momentum states above the Hund's rule ground state. Therefore, a least squares fit of the data above 0.87 K was made to the function

$$\chi_M = \frac{N_A}{k_B}\left[\frac{\mu_{eff}^2}{3(T-\theta_p)} + \frac{\mu_B^2}{\delta}\right] , \qquad (1)$$

where N_A is Avogadro's number, k_B is Boltz-

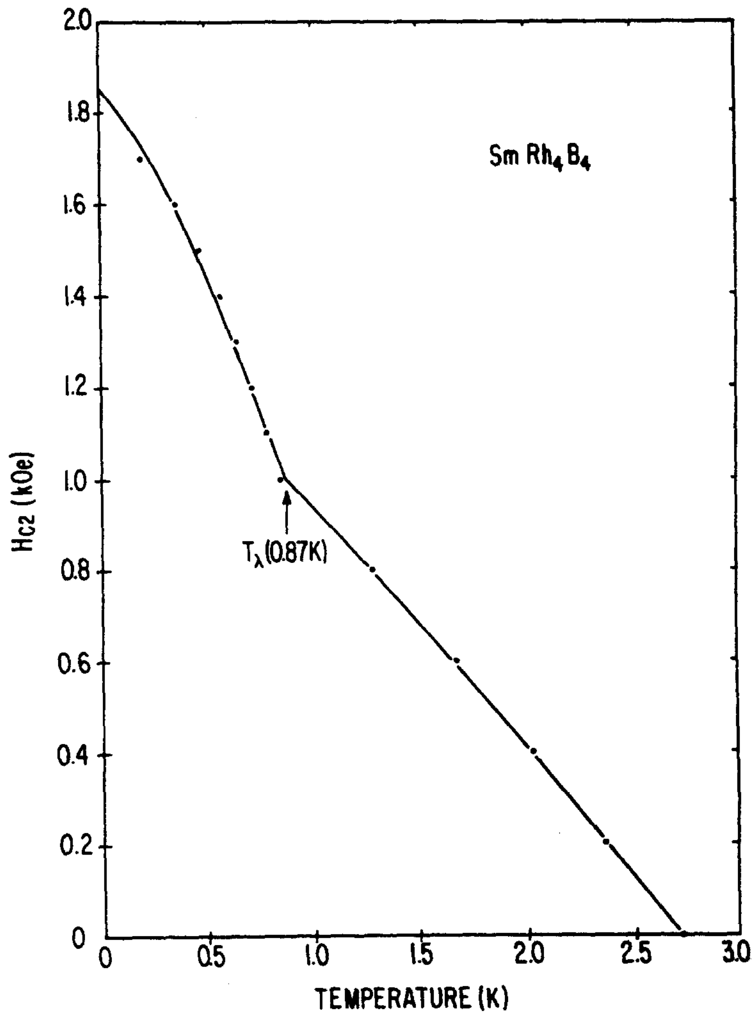

Fig. 2 Upper critical field H_{c2} versus temperature for $SmRh_4B_4$. The arrow represents the transition temperature defined by the lambda-type anomaly observed in the heat capacity data. The curve represents a fit of the data to Machida's theory[8] for antiferromagnetic superconductors.

mann's constant, μ_B is the Bohr magneton, μ_{eff} is the effective magnetic moment, and θ_p is the Curie-Weiss temperature.[6] The first term represents a Curie-Weiss contribution from the $J = 5/2$ ground state, while the second term is the temperature independent Van Vleck correction arising from the accessible first excited angular momentum $J = 7/2$ state. In the absence of crystal field effects, $\mu_{eff} = g_J\sqrt{J(J+1)}\mu_B = 0.845\ \mu_B$, where g_J is the Landé g-factor, and $\delta = 7\ \Delta E/20$, where ΔE is the difference in energy between the $J = 5/2$ and $J = 7/2$ angular momentum states. Using the best fit parameters of $\mu_{eff} = 0.632\ \mu_B$, $\theta_p = -1.93$ K, and $\delta = 377$ K, Eq. (1) describes the data reasonably well, as shown in Fig. 4. However, the effective magnetic moment $\mu_{eff} = 0.632\ \mu_B$ is considerably less than the $0.845\ \mu_B$ free ion value for the $J = 5/2$ Hund's rule ground state of Sm^{3+}, while $\delta = 377$ K corresponds to $\Delta E = 1080$ K, a value somewhat less than but comparable to the ~ 1500 K value estimated for free Sm^{3+} ions.[7] In contrast, the magnetic susceptibilities of $ErRh_4B_4$ [3] and $NdRh_4B_4$ [5] yielded effective magnetic moments nearly equal to their free ion values. The re-

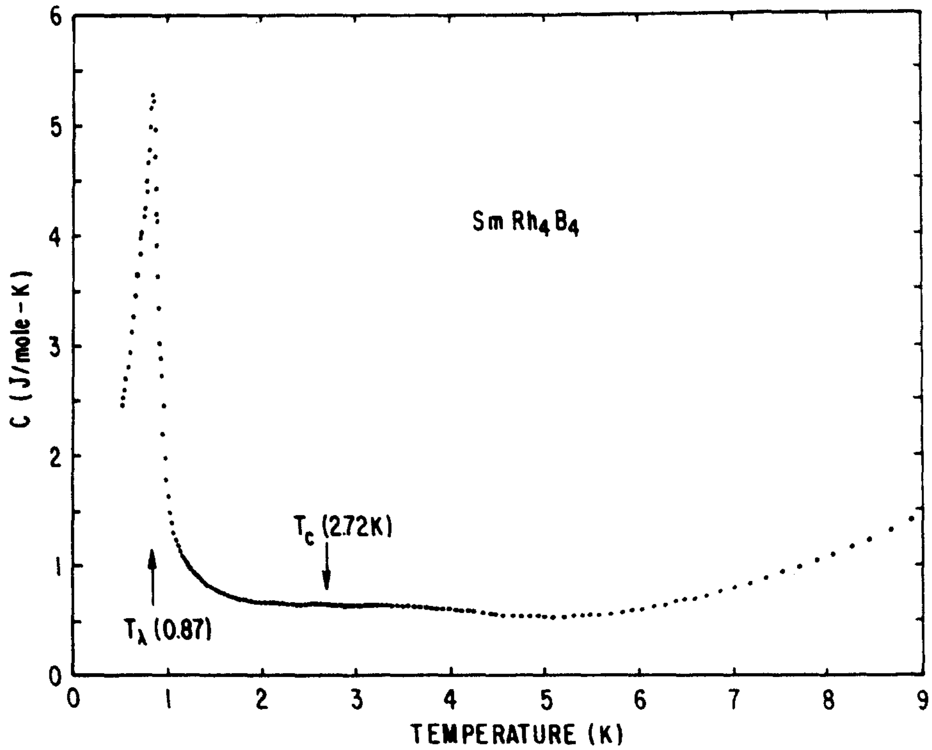

Fig. 3 Specific heat C versus temperature for $SmRh_4B_4$ in zero applied magnetic field.

duced value of μ_{eff} for $SmRh_4B_4$ is consistent with the specific heat data which revealed crystal field splitting of the Hund's rule ground state energy levels. The magnetic susceptibility curve also exhibits a cusp-like feature near T_λ, indicative of an antiferromagnetic transition at this temperature.

The ac electrical resistance, upper critical field, specific heat, and magnetic susceptibility data all indicate that $SmRh_4B_4$ orders antiferromagnetically in the superconducting state at $T_\lambda = 0.87$ K. This immediately suggests that the H_{c2} vs temperature data for $SmRh_4B_4$ may provide a useful test of theories of antiferromagnetic superconductors, such as the one recently developed by Machida.[8] In Machida's model, which is an extension of the Abrikosov-Gor'kov theory,[9] the equation relating the upper critical field and the temperature is given by

$$\ln\frac{T}{T_{co}} + \frac{1}{2}\left[\left(1+\frac{b}{\sqrt{b^2-h^2}}\right)\psi\left(\frac{1}{2}+\rho_-\right) + \left(1-\frac{b}{\sqrt{b^2-h^2}}\right)\psi\left(\frac{1}{2}+\rho_+\right)\right] - \psi\left(\frac{1}{2}\right) = 0\,, \quad (2)$$

where

$$\rho_\pm = \frac{1}{2\pi T}\left(\frac{1}{\tau(T)} + b + DeB \pm \sqrt{b^2-h^2}\right) \quad (3)$$

and

$$h = \frac{g_J\mu_B\, IS_o(T)}{3k_BT}H + \mu_B B\,. \quad (4)$$

Here ψ is the digamma function, T_{co} is the critical temperature in the absence of ions that carry magnetic moments, $b = 1/\tau_{so}$ where τ_{so} is the spin-orbit relaxation time, $D = \tau_o v_f^2/3$ is a diffusion constant where v_f is the Fermi velocity and τ_o is the relaxation time associated with scattering of the conduction electrons by nonmagnetic impurities, e is electronic charge, I is the exchange integral characterizing the strength of the interaction between the local moments and the conduction electrons, $\tau(T)$ is the temperature dependent relaxation time of the conduction electron system, and S_o is the $q=0$ value for the spin-spin correlation function $S_q = \langle \vec{J}_q \cdot \vec{J}_{-q}\rangle$. The relation between S_o and τ is given by

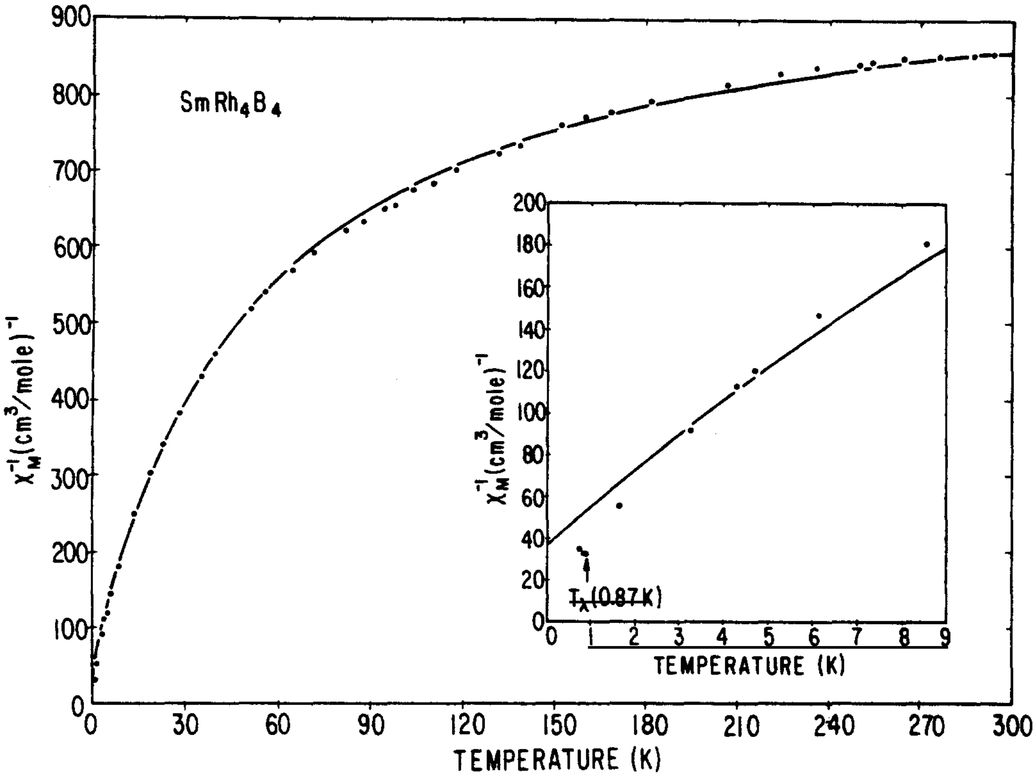

Fig. 4 Inverse magnetic susceptibility per mole χ_M^{-1} versus temperature for $SmRh_4B_4$. The arrow represents the transition temperature defined by the lambda-type anomaly observed in the heat capacity data. The curve represents the sum of a Curie-Weiss law with μ_{eff} = 0.632 and θ_p = -1.93 K and a temperature independent Van Vleck term.

$$\frac{1}{\tau(T)} = 2\pi N(E_F)\left(\frac{I}{2}\right)^2 (g_J - 1)^2 S_o(T), \quad (5)$$

where $N(E_F)$ is the density of states at the Fermi level, while $S_o(T)$ is related to the magnetic susceptibility χ by the expression

$$\chi = N \frac{g_J^2 \mu_B^2}{3k_B T} S_o, \quad (6)$$

where N is the number of magnetic moments per unit volume. For χ the standard mean field theory expression[10] for an isotropic, polycrystalline antiferromagnetic material was adopted using the Hund's rule free ion values for g_J and J and assuming that both the Neél temperature and the magnitude of the Curie-Weiss temperature were equal to T_λ. Although the magnetic susceptibility data seem to contradict these assumptions, this simplified model provides a qualitative description of the behavior of χ at the temperatures $T \leq 2.7$ K of interest here. The value used for $N(E_F)$ was 0.57 states/eV-atom-spin direction, the density of states at the Fermi level for the isostructural nonmagnetic compound $LuRh_4B_4$,[11] while N was calculated from the crystallographic data.[2]

The results of the numerical fit are shown in Fig. 2, with the optimum value of the parameters being $1/\tau_{so}$ = 20.15 K, De/μ_B = 32.33, T_{co} = 4.95 K, and I = 8.58 × 10^{-3} eV-atom. The agreement between experiment and theory is excellent, especially in view of the simplifications discussed above. The enhancement of the upper critical field at low temperatures therefore appears to be the result of the reduction of the net magnetization below T_λ. The value for I obtained here is roughly comparable to the value 4×10^{-2} eV-atom that was previously inferred from the rate of the depression of T_c with x in the pseudoternary system $(Lu_{1-x}Ho_x)Rh_4B_4$.[12] Also of interest is the small value of T_{co} in comparison to the critical temperature of 11.6 K for $LuRh_4B_4$. If reliable, this significantly reduced value of T_c in the absence of magnetic pairbreaking interactions suggests that the superconducting transition temperature of the tetragonal $RERh_4B_4$ compounds depends sensitively on the lattice parameters.

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REFERENCES

1. MATTHIAS, B. T., CORENZWIT, E., VANDENBERG, J. M. and BARZ, H. E., Proceedings of the National Academy of Sciences USA 74, 1334 (1977).
2. VANDENBERG, J. M. and MATTHIAS, B. T., Proceedings of the National Academy of Sciences USA 74, 1336 (1977).
3. FERTIG, W. A., JOHNSTON, D. C., DeLONG, L. E., McCALLUM, R. W., MAPLE, M. B. and MATTHIAS, B. T., Physical Review Letters 38, 987 (1977).
4. MONCTON, D. E., McWHAN, D. B., ECKERT, J., SHIRANE, G. and THOMLINSON, W., Physical Review Letters 39, 1164 (1977).
5. HAMAKER, H. C., WOOLF, L. D., MacKAY, H. B., FISK, Z. and MAPLE, M. B., to appear in Solid State Communications.
6. See, for example, WAGNER, D., Introduction to the Theory of Magnetism, Pergamon Press, 1972, pp. 94-99.
7. VAN VLECK, J. H., The Theory of Electric and Magnetic Susceptibilities, Oxford Press, 1932, pp. 245-256.
8. MACHIDA, K., to be published.
9. ABRIKOSOV, A. A. and GOR'KOV, L. P., JETP 12, 1243 (1961).
10. WAGNER, D., Introduction to the Theory of Magnetism, Pergamon Press, 1972, pp. 94-99.
11. WOOLF, L. D., JOHNSTON, D. C., MacKAY, H. B., McCALLUM, R. W. and MAPLE, M. B., Journal of Low Temperature Physics 35, 651 (1978).
12. MAPLE, M. B., HAMAKER, H. C., JOHNSTON, D. C., MacKAY, H. B. and WOOLF, L. D., Journal of Less Common Metals 62, 251 (1978).