UC Irvine
UC Irvine Previously Published Works

Title

Superconductivity and magnetism in ternary rare-earth compounds

Permalink

https://escholarship.org/uc/item/07b9f3kx

Journal

Physica B+C, 120(1-3)

ISSN

0378-4363

Authors

Mook, HA
Pringle, OA
Kawarazaki, S
et al.

Publication Date

1983-05-01

DOI

10.1016/0378-4363(83)90372-8

§2.3. SPIN DENSITY DISTRIBUTIONS

SUPERCONDUCTIVITY AND MAGNETISM IN TERNARY RARE-EARTH COMPOUNDS

H.A. MOOK and O.A. PRINGLE

Oak Ridge National Laboratory[1], Oak Ridge, TN 37830, USA

S. KAWARAZAKI[2]

Osaka University, Osaka 560, Japan

S.K. SINHA, G.W. CRABTREE and D.G. HINKS

Argonne National Laboratory[3], Argonne, IL 60439, USA

M.B. MAPLE[4], Z. FISK, D.C. JOHNSTON[6], L.D. WOOLF[4,5] and H.C. HAMAKER[7]

University of California, San Diego, La Jolla, CA 92093, USA

Invited paper

Superconductivity and magnetism are two types of order that can take place in materials at low temperatures. When the magnetic order is ferromagnetic, a competition exists between magnetism and superconductivity. Neutron scattering has been used to measure the interaction of magnetism and superconductivity in a series of ternary rare-earth alloys. A wide range of behavior is found near the magnetic transition, including mean-field magnetic ordering, first-order transitions between magnetism and superconductivity, and co-existence of ferromagnetism and superconductivity with a sinusoidally-modulated magnetic phase.

When a material is cooled to low temperatures magnetism and superconductivity are two types of long range order that can take place. When the magnetic order is ferromagnetic so that a net long range magnetic field is established, a competition develops between the magnetic and superconducting state. The discovery of reentrant superconductors such as $ErRh_4B_4$ [1] and $Ho_{1.2}Mo_6S_8$ [2] has given us an interesting testing ground for the study of the interaction of superconductivity and long range ferromagnetism, and a number of interesting experiments have been performed on these materials. Considerable theoretical work has also been done in order to understand the interaction between magnetism and superconductivity in these types of materials. A review of the more recent work is given in ref. [3].

Neutron scattering is a valuable probe for the study of magnetism in materials and several neutron scattering investigations have been performed on materials exhibiting reentrant superconductivity. One of the most interesting systems is the $Ho_{1-x}Er_xRh_4B_4$ pseudoternary alloy system, since a wide range of phenomena occurs throughout this system, ranging from pure mag-

netism on the Ho rich end to coexistence of superconductivity and magnetism for ErRh₄B₄. We will also mention a few recent measurements made on the Ho(Rh₁₋ₓIrₓ)₄B₄ system where additional interesting behavior is found.

The magnetic neutron scattering from magnetic ions in these materials is given by

$$I \sim |F_M|^2 = 4\left(\frac{\gamma e^2}{2mc^2}\right)^2 \mu^2 f(K)^2 [1 - (\hat{K} \cdot \hat{\mu})^2] , \qquad (1)$$

where $\gamma e^2/2mc^2 = -0.2695 \times 10^{-12}$ cm, μ is the magnetic moment, K is the neutron momentum transferred to the crystal, $\hat{\mu}$ is the moment direction and $f(K)$ is the magnetic form factor. $f(K)$ is not known for these compounds but is probably similar to that observed for Ho and Er in other rare-earth compounds. Most of our measurements are made at a fixed K, so that $f(K)$ is a constant in any case. We can determine the direction of magnetic ordering by the $1 - (\hat{K} \cdot \hat{\mu})^2$ term, and the width of the diffraction peaks from crystals or powders gives us a lower limit on the spatial extent of the magnetic ordering. Only powders of these materials are available, except for ErRh₄B₄, where a single crystal is available.

Quite a lot of information can be obtained from a measurement of the magnetic moment μ as a function of temperature. For a powder sample, the neutron intensity I for a given reflection is proportional to μ^2, so that I vs. temperature gives the magnetic order parameter. Fig. 1 shows the phase diagram of the (Er, Ho)Rh₄B₄ [4] system. The arrows show compositions that have been investigated by neutron scattering. The end point material HoRh₄B₄ does not go superconducting, so measurements on this material give an indication of what the magnetic order should be like in the absence of superconductivity. Fig. 2 shows the magnetic order parameter for HoRh₄B₄ as determined from the (101) reflection. The ordering in this material is of a particularly simple nature in that a spin 1/2 mean-field model gives a good account of the shape of the magnetic order parameter. This mean-field ordering has been discussed in detail by Ott et al. [5], and the line through the points
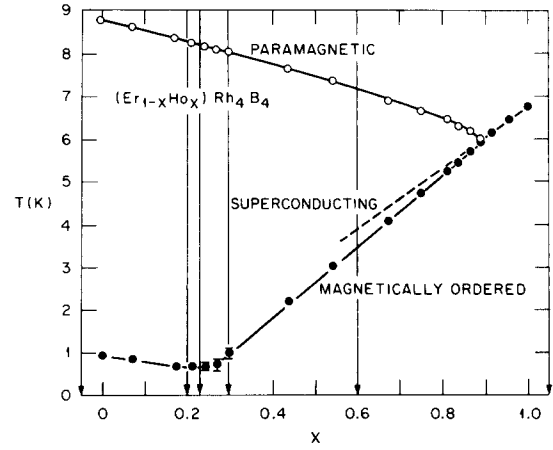

Fig. 1. Phase diagram for the (Er, Ho)Rh₄B₄ pseudoternary alloy system.

is given by their equation for a mean-field model of a magnet. Note that a well-defined transition is seen at 6.70 K with no evidence of critical scattering above the transition. This type of simple ordering is quite unusual as there is usually rounding near the transition by critical fluctuations. The resolution of the powder diffractometer used was such that it could be established that the order was longer than 200 Å, and the magnetic moment was found to be along the c-axis.
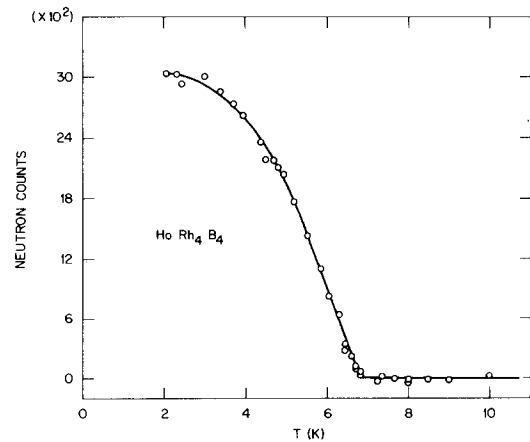
As ErRh₄B₄ is added to HoRh₄B₄ the situation

Fig. 2. Magnetic intensity vs. temperature for the (101) reflection of HoRh₄B₄. The solid line is given by a simple spin 1/2 mean-field model.

becomes more complicated. The point where superconductivity and magnetism join at about the composition $Ho_{0.9}Er_{0.1}Rh_4B_4$ probably has complicated behavior, and Grewe and Schuh [6] have suggested that Lifshitz point behavior may be found near this composition. We have not investigated this region but have made careful measurements at the composition $Ho_{0.6}Er_{0.4}Rh_4B_4$ [7], which is a reentrant superconductor. It is found that superconductivity changes the mean-field transition into a first-order transition. The order parameter curve looks mean-field-like until the region near the transition, where a sudden deviation is made from the mean-field curve. Woolf et al. [8] have analyzed the neutron scattering results to show that the magnetic ordering temperature is depressed by about 0.2 K because of the occurrence of superconductivity. Magnetism and superconductivity are separated by the sharp first-order transition and no coexistence region is found. The ordered moment is about the value we would expect if only the Ho ordered with its free ion value. Probably the Er orders at a much lower temperature, if indeed it orders at all. The direction of the ordering is again along the c-axis.

As one adds more Er into the alloy system, a minimum is found in the magnetic ordering temperatures at compositions around $Er_{0.3}Ho_{0.7}Rh_4B_4$. As this point is approached from the Ho rich side an additional ordering, corresponding to basal plane ordering, is found at a lower temperature. Finally, as alloys are investigated on the Er rich side of the minimum, it is found that basal plane ordering takes place at a higher temperature and c-axis ordering occurs at a lower temperature. $ErRh_4B_4$ is found to have basal plane ordering with a moment of about $5.5\mu_B$ [9]. Destruction of superconductivity takes place in a sharp transition upon c-axis ordering, but remains to some extent when the basal plane orders first. The order parameter for the basal plane looks very different than for the compositions where c-axis ordering occurs, in that it has a long tail that extends to high temperatures.

The order parameter for $ErRh_4B_4$ is shown in fig. 3 along with the satellites that accompany the
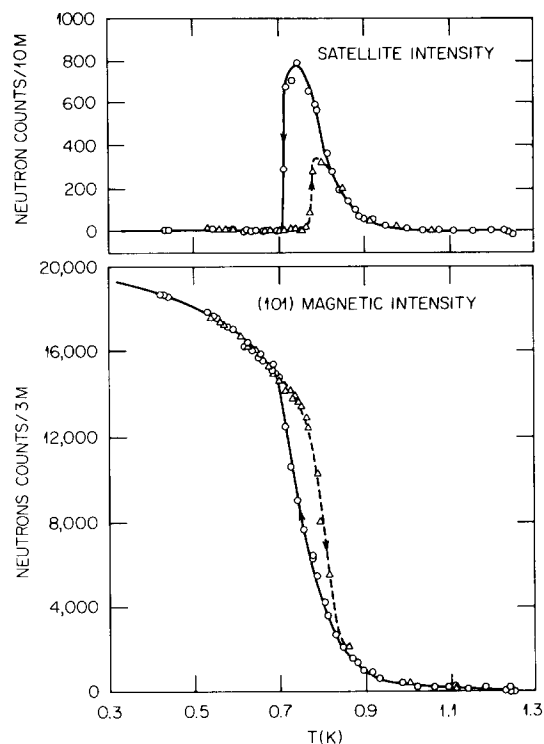

Fig. 3. Magnetic intensity vs. temperature for the (101) reflection of a single crystal of $ErRh_4B_4$. The intensity of one of the satellites is also shown.

main Bragg peaks. This measurement was made on a small single crystal of the material. Earlier measurements on powders by Moncton et al. [10] showed peaks in the small angle scattering that suggested an oscillatory state was present near the transition, confirming the theoretical predictions of Blount and Varma [11]. The single crystal measurements showed that the small angle peaks resulted from satellites around the Bragg peaks. The satellite positions and intensities indicate that a linearly polarized sinusoidal modulation of about 90 Å occurs in the magnetic moment in the temperature region where the satellites are found [12]. On cooling, superconductivity remains until the satellites disappear at about 0.7 K, at which point only basal plane ferromagnetic order is found. Upon warming, the satellites and superconductivity are regained at about 0.74 K. There is thus a region where ferromagnetism, superconductivity, and a modu-

lated moment coexist. The shape of the (101) magnetic intensity near the ordering temperature shows that ordering does not take place in the usual manner, and it seems likely that small ferromagnetic domains grow slowly from the superconductor as the temperature is lowered, while other domains remain superconducting but have a modulated moment as well. Fig. 4 shows the difference between a scan taken at 2.73 K, which is above the ordering temperature, and a scan taken at 1.1 K, which is on the tail of the order parameter. The nuclear intensity is large compared to the magnetic intensity for the (101) peak at 1.1 K, so it is hard to get good counting statistics. Nevertheless, the difference, which is the purely magnetic scattering, is considerably broadened from the nuclear-only scattering at 2.73 K, showing that the order is not very long
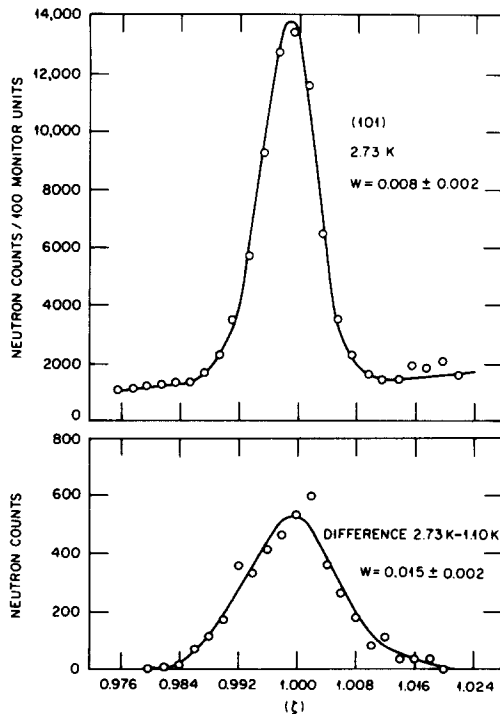

Fig. 4. Scan of the (101) Bragg peak for $ErRh_4B_4$ in units of $(\zeta 0\zeta)$. At 2.73 K the scattering is nuclear only, showing a resolution width of about 0.008 in ζ. The lower curve shows the difference between the (101) scans at 2.73 and 1.1 K. This difference gives the magnetic intensity. The scan at 1.1 K is seen to be wider, thus showing the finite range of magnetic order as it is first established.

range in this temperature region. This is what would be expected if the magnetization grew from small domains in the superconductor. High resolution measurements at 0.8 K show long range order of at least 10 000 Å, so the domains have grown large by this temperature. Tachiki [13] has made a model calculation for the domain structure in the coexistence region, but it seems that his calculations suggest domains smaller than 10 000 Å. Bulaevskii et al. [14] have suggested a domain model for the coexistent state which consists of domains where magnetization alternates along a planar array. This would require satellites to occur at $(2M + 1)$ positions in a^* and c^*. However, we have checked that no $3a^*$, c^* satellite exists, thus ruling out such a model.

It would be very interesting if additional information could be obtained about the nature of the coexistent state. We will try further measurements near the transition to see if a better understanding can be gained about how the magnetically ordered regions establish themselves.

Other reentrant superconductors are of interest as well. Yang et al. [15] have mapped out the phase diagram for the $Ho(Rh_{1-x}Ir_x)_4B_4$ system. We have made measurements on $Ho(Rh_{0.85}Ir_{0.15})_4B_4$ and $Ho(Rh_{0.3}Ir_{0.7})_4B_4$. Behavior similar to the high Ho concentration alloys in the $(Ho, Er)Rh_4B_4$ system is found for $Ho(Rh_{0.3}Ir_{0.7})_4B$, except that some deviations from mean-field behavior are found. $Ho(Rh_{0.3}Ir_{0.7})_4B_4$ is found to be simultaneously antiferromagnetic and superconducting with the magnetic order occurring first at 2.7 K and then superconductivity at 1.42 K. The magnetic structure consists of stacked antiferromagnetic basal plane sheets forming a body-centered tetragonal unit cell. For compositions intermediate to the two studied, the phase diagram is very complicated and further neutron scattering measurements are needed to understand the magnetic behavior.

Alloy series of the above type contain materials with many different types of magnetic and superconducting behavior. The balance between magnetism and superconductivity is a delicate

one, so that small changes in alloy composition make different phenomena occur. The materials then are a good testing ground in which to study magnetism and superconductivity and to see how they interact.

References

[1] W.A. Fertig, D.C. Johnston, L.E. DeLong, R.W. McCallum, M.B. Maple and B.T. Matthias, Phys. Rev. Lett. 38 (1977) 987.

[2] M. Ishikawa and Ø. Fischer, Solid State Commun. 23 (1977) 37.

[3] H.A. Mook, W.C. Koehler, M.B. Maple, Z. Fisk, D.C. Johnston and L.D. Woolf, Phys. Rev. B25 (1982) 372.

[4] D.C. Johnston, W.A. Fertig, M.B. Maple and B.T. Matthias, Solid State Commun. 26 (1978) 141.

[5] H.R. Ott, G. Keller, W. Odoni, L.D. Woolf, M.B. Maple, D.C. Johnston and H.A. Mook, Phys. Rev. B25 (1982) 499.

[6] N. Grew and B. Schuh, in: Proc. of the IV Conf. on Superconductivity in d- and f-Band Metals, Karlsruhe, Germany (1982).

[7] H.A. Mook, W.C. Koehler, S.K. Sinha, G.W. Crabtree, D.G. Hinks, M.B. Maple, Z. Fisk, D.C. Johnston, L.D. Woolf and H.C. Hamaker, J. Appl. Phys. 53 (1982) 2614.

[8] L.D. Woolf, D.C. Johnston, H.A. Mook, W.C. Koehler, M.B. Maple and Z. Fisk, in: Proc. of the Low Temperature Conference – LT-16 (1982). Physica 109 & 110B (1982) 2045.

[9] D.E. Moncton, D.B. McWhan, J. Eckert, G. Shirane and W. Thomlinson, Phys. Rev. Lett. 39 (1977) 1164.

[10] D.E. Moncton, D.B. McWhan, P.H. Schmidt, G. Shirane, W. Thomlinson, M.B. Maple, H.B. MacKay, L.D. Woolf, Z. Fisk and D.O. Johnston, Phys. Rev. Lett. 45 (1980) 2060.

[11] E.I. Blount and C.M. Varma, Phys. Rev. Lett. 42 (1979) 1079.

[12] S.K. Sinha, G.W. Crabtree, D.G. Hinks and H.A. Mook, Phys. Rev. Lett. 48 (1982) 950.

[13] M. Tachiki, in: Proc. of the IV Conf. on Superconductivity in d- and f-Band Metals, Karlsruhe, Germany (1982).

[14] L.N. Bulaevskii, A.I. Buzdin and M.L. Kulic, in: Proc. of the IV Conf. on Superconductivity in d- and f-Band Metals, Karlsruhe, Germany (1982).

[15] K.N. Yang, S.E. Lambert, H.C. Hamaker, M.B. Maple and H.A. Mook, in: Proc. of the IV Conf. on Superconductivity in d- and f-Band Metals, Karlsruhe, Germany (1982).