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MAGNETIC FIELD DEPENDENCE OF THE NEUTRON SCATTERING FROM  $\text{ErRh}_4\text{B}_4$  \*

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Magnetism in the reentrant superconductor  $\text{ErRh}_4\text{B}_4$  has been studied by neutron scattering as a function of an applied magnetic field. For a temperature of 1.69 K long-range ferromagnetism is found in fields higher than 1 kOe. Considerable hysteresis is found in the neutron scattering intensity vs magnetic field curve and long-range order with a small Er moment remains when the field is reduced to small values.

BECAUSE of its unusual magnetic and superconducting properties [1] there has been a lot of interest in the ternary compound  $\text{ErRh}_4\text{B}_4$ . The material becomes a superconductor at 8.7 K and superconductivity is destroyed and long-range magnetic order is established at about 0.9 K. Neutron diffraction experiments by Moncton *et al.* [2] have shown that  $\text{ErRh}_4\text{B}_4$  is a ferromagnet below 0.9 K with the moment direction in the basal plane. The moment value was found to be  $5.6 \mu_B$  which is well below the free ion value of  $9.0 \mu_B$ . Fertig *et al.* [1] showed that the superconducting state is destroyed with the application of a magnetic field. This paper reports neutron diffraction measurements on  $\text{ErRh}_4\text{B}_4$  taken as a function of an applied static magnetic field.

The sample was prepared by arc melting the rare earth tetraboride with Rh followed by annealing. The  $^{11}\text{B}$  isotope was used to decrease the absorption cross section for slow neutrons. The experiments were performed at the High Flux Isotope Reactor using conventional techniques. An incident neutron wavelength of 2.46 Å was used so that pyrolytic graphite could be

used to remove high order contamination from the monochromator. The monochromator was pyrolytic graphite and collimation after the sample of  $20'$  was used. Two sets of measurements were made. In the first set of measurements the sample was placed in a superconducting magnet capable of producing high magnetic fields. However, the superconducting magnet had a small remanent field and thus it was necessary to check the low field results with a different magnet system. Additional low field measurements were thus made with a conventional pumped  $^4\text{He}$  cryostat placed in a Helmholtz pair of magnet coils. This gave a uniform magnetic field and a negligible remanent field. The experiments were performed at the lowest temperatures achievable by the two cryostat systems which were 1.69 K for the superconducting magnet and 1.79 K for the cryostat with the Helmholtz coils.

We will first discuss the measurements made in the superconducting magnet. Magnetic diffraction peaks could easily be observed in fields larger than about 1 kOe. The diffraction pattern was identical to the one obtained by Moncton *et al.* [2] at low temperatures with no applied field. The field thus destroys the superconductivity but does not alter the magnetic structure from that found in the low temperature phase in zero field. Figure 1 shows the (101), (110) and (002) peaks taken with no applied field and in an applied field of 10 kOe. The small peaks found in zero field are attributable to nuclear scattering only and indicate that there is no long-range magnetic order. The peaks found at 10 kOe show ferromagnetic ordering with the moment oriented in the tetragonal basal plane. The peaks are resolution limited showing long-range magnetic order of at least 150 Å.

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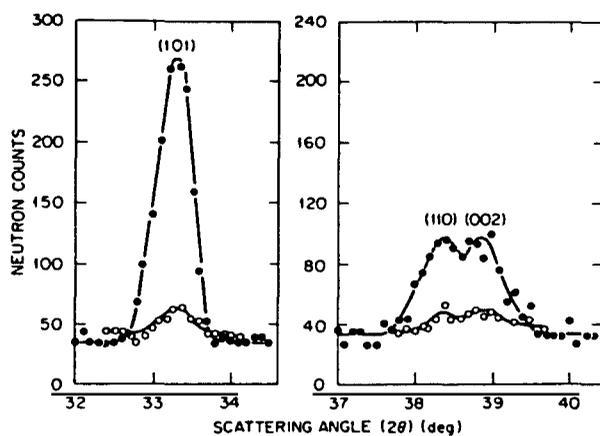


Fig. 1. Powder diffraction peaks from  $\text{ErRh}_4\text{B}_4$  at zero applied field (open circles) and in an applied field of 10 kOe (closed circles).

When making powder diffraction measurements in a magnetic field considerable care must be taken that the field does not orient the powder particles preferentially. This commonly occurs with fine particles that have sizeable magnetic moments. In the present case, the sample consisted of rather coarse polycrystalline particles held in a flat sample holder so that the sample size was about  $2 \times 2 \times 0.1 \text{ cm}^3$ . The field was applied vertically so that it was perpendicular to the scattering plane and along one of the long dimensions of the plate. The thin plate was used so that neutrons could be transmitted through the sample which is quite absorbing to slow neutrons because of the high absorption cross-section of Rh. The sample particles were packed tightly in the holder and it seems unlikely that they would move in the presence of the applied field. Nevertheless, to check that the field was not producing preferred orientations that could influence the interpretation of our results, powder diffraction patterns were taken before and after the fields were applied and at several values of the applied field. Analysis of the diffraction patterns showed no evidence that the field was producing preferential alignment of a particular crystallographic axis.

The field dependence of the magnetic scattering determined from the intensity of the (101) reflection is shown in Fig. 2. The nuclear component to the reflection has been subtracted. The field was produced by the superconducting magnet operating in the persistent mode and thus the field value was very stable during the measurements at each point. The sample temperature was held at  $1.69 \pm 0.02 \text{ K}$  during the course of the experiment. The curve for increasing field is in some respects similar to that established by Fertig *et al.* [1] and Ott *et al.* [3, 4] by magnetization measurements for increasing fields. No intensity is observed until a field of

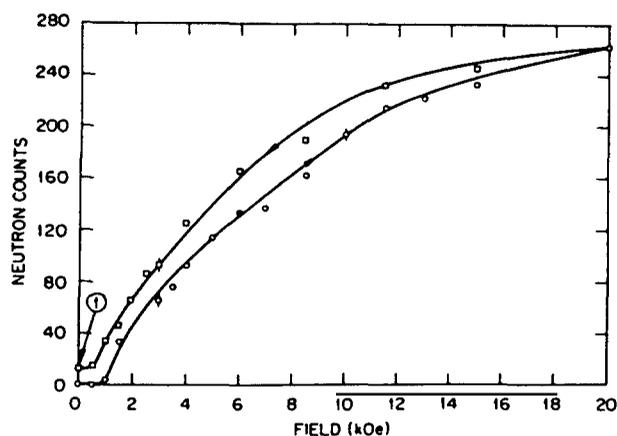


Fig. 2. Field dependence of the magnetic contribution to the (101) peak intensity measured in a superconducting magnet for a sample temperature of 1.69 K.

about 1 kOe was applied reflecting the superconductivity in this field range. The scattering intensity then increases with increasing field, the ordered moment values corresponding to  $5.06 \pm 0.5 \mu_B$  at 10 kOe and  $6.9 \pm 0.5 \mu_B$  at 20 kOe. This is a similar value to that obtained in the magnetization measurements. The intensity found upon lowering the applied field appears to be quite different from the magnetization measurements although the reversible magnetization data are only published for low field values. We see considerable hysteresis in the intensity vs field curve and at zero applied field we still see some long-range ordered moment. The field value is not brought identically to zero since the magnet assembly and spectrometer have some remanent field amounting to about 100 Oe.

Good statistics were obtained after decreasing the field to zero applied field (point 1 on the graph) showing that the long-range order found is a real effect. If one starts from point 1 and increases the field, the intensity remains on the decreasing field curve and follows it back to higher values. If one starts at point 1 and warms the sample to a temperature above the superconducting transition temperature (8.7 K) and then recools to 1.69 K one returns to the zero intensity value corresponding to no long-range moment. The scattering intensity at point 1 corresponds to a long-range ordered moment of  $0.7 \pm 0.2 \mu_B$ . Freeman and Jarlborg [5] have in fact previously suggested that upon lowering an external field from a value greater than the critical field it may be possible to form a mixed state in which normal and ferromagnetic regions of the compound coexist with superconductivity.

The results of the second set of measurements are shown in Fig. 3. These measurements are confined to low field values but are free of any effects that might be caused by any remanent field. Upon increasing the

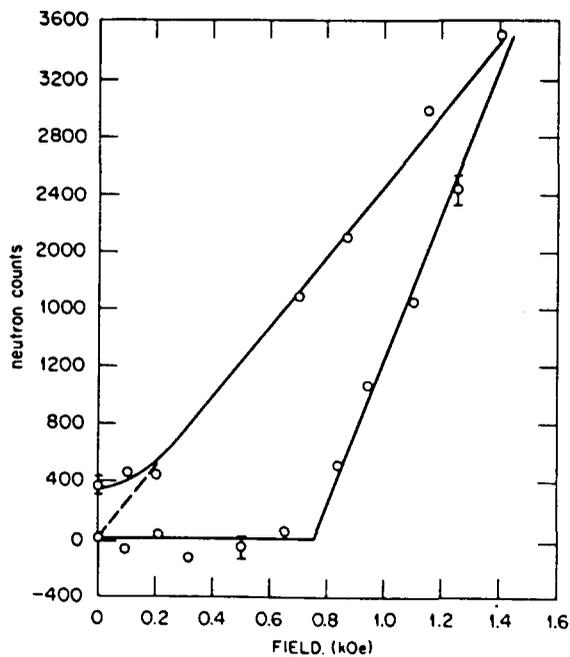


Fig. 3. Field dependence of the magnetic contribution to the (101) peak intensity measured in a Helmholtz pair of coils for a sample temperature of 1.79 K.

field one sees a similar result as was obtained in the first set of measurements. No long-range order is found for fields smaller than about 0.7 kOe. For fields above this value a long-range ordered moment develops rapidly. Very little precursor scattering is found at fields lower than the critical field value for developing long-range order. This is unlike the manner in which long-range order is developed by cooling through the transition where considerable precursor scattering is found at temperatures above the transition.

As the field is increased above 0.8 kOe the long-range ordered moment increases with increasing field in a nearly linear manner. At 1.5 kOe which was the highest field attainable in the Helmholtz coils the field was reduced in value. The long-range ordered moment decreases with decreasing field in a linear fashion but with a slope that extrapolates through the origin. As one reaches small field values the curve departs from the linear slope and one again finds a long-range ordered

moment at zero field. Upon heating the sample above 10 K and returning to 1.79 K one finds again that there is no long-range order in zero applied field.

The two sets of measurements thus show similar effects. No long-range order is found for values less than a critical field value but the long-range ordered moment develops rapidly with increasing field values once the critical field is attained. The ordered moment develops in a manner similar to that given by magnetization measurements. As the field is lowered the long range moment decreases with decreasing field but no reversible region is found in fields up to 20 kOe. For low fields the ordered moment decreases on a curve that extrapolates to zero but departs from this curve for fields under about 0.2 kOe. A small long range ordered moment remains at zero field but disappears upon warming above the superconducting transition temperature and returning to low temperatures.

The difference between the neutron and magnetization measurements reflect the fact that only the long-range ordered moment is visible in the neutron experiment while the magnetization measurements sample all of the moment independently of whether or not it has long-range order. Some of this moment may exist as trapped flux in a fluxoid lattice. The fluxoid spacings are usually on the order of 1000 Å and would not be directly visible in the present neutron scattering experiment. Experiments are planned with sufficient resolution to directly observe any fluxoid lattice that might be formed.

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