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LETTER TO THE EDITOR

Strong itinerant magnetism in ternary boride CeRh_3B_2

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Abstract. We report in this Letter a high-temperature itinerant ferromagnet CeRh_3B_2 which orders magnetically at 115 K. The magnetic ordering seems to be intrinsic and presumably arises from the Rh d-band. Similar compounds of La, Pr and Nd do not show any magnetic ordering down to 77 K.

The ternary borides containing rare earths have been of much current interest. Many of these compounds are superconducting; some are magnetically ordered while a few others show the re-entrant behaviour and even coexistence of superconductivity and magnetic ordering (Matthias *et al* 1977, Fertig *et al* 1977, Hamaker *et al* 1979a, b, Ku *et al* 1979). This has provoked a great deal of experimental and theoretical work in this field.

Recently, a new series of ternary borides having the formula RT_3B_2 has been reported (Ku *et al* 1980). Here R stands for the rare earth ion, and T is one of the elements belonging to the 4d (Ru and Rh) or 5d (Os and Ir) series. These compounds have the hexagonal CeCo_3B_2 type structure, which in turn is derived from the binary CaCu_5 type structure, space group $P6/mmm$ (D_{6h}^1). There is only one formula unit of RT_3B_2 per unit cell and a unique site for the rare earth ion. Details of the structure and the rare earths which form RT_3B_2 compounds are given by Ku *et al* (1980). The RT_3B_2 compounds show superconducting behaviour with T_c up to 4.6 K for rare earths which do not carry a magnetic moment, e.g. La and Lu and also Th. For compounds where R is a moment-carrying rare earth ion, magnetic ordering is observed with Curie temperatures sometimes exceeding 45 K. Examination of the volume per unit cell of RRh_3B_2 and RRu_3B_2 compounds (Ku *et al* 1980) reveals an anomaly in the case of cerium compounds which indicates that cerium differs from its normal trivalent state. Because of the long standing interest in our laboratory in compounds containing cerium (Malik *et al* 1974, 1975) and other rare earths capable of showing abnormal valencies, it was decided to examine more carefully the magnetic behaviour of CeRh_3B_2 and CeRu_3B_2 . We find that the compound CeRh_3B_2 is magnetically ordered with a relatively high Curie temperature of 115 K. From the low magnetic moment per formula unit, it appears that the ordering is of the itinerant type and is very likely associated with the 4d band of Rh metal. To our knowledge this is the highest reported T_c in any cerium compound with non-magnetic elements. No magnetic ordering is observed in CeRu_3B_2 down to 77 K.

The intermetallic compounds were prepared by melting together stoichiometric amounts of the constituent elements in an argon arc furnace. The alloy buttons were

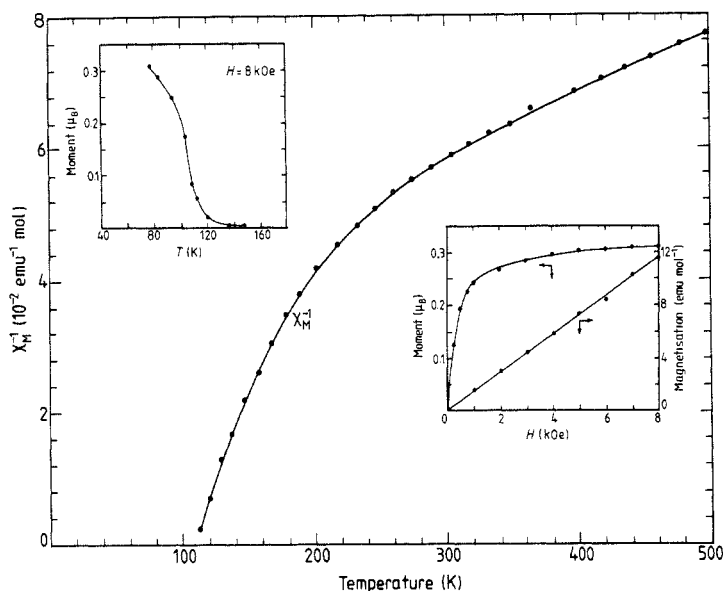


Figure 1. Temperature dependence of inverse molar susceptibility of CeRh_3B_2 . The top inset shows the magnetisation in an applied field of 8 kOe near T_c . The bottom inset shows magnetisation against applied field at 77 K (top curve) and 300 K (bottom curve).

turned and melted several times to obtain thorough mixing. The purity of the starting materials was as follows: rare earths 99.9%; Rh and Ru 99.95%; and boron obtained from Koch Light had a purity of 99.99%. The elements Rh, Ru and B were checked for any magnetic impurity by magnetic susceptibility measurements. Powder x-ray diffraction patterns of the compounds were obtained using $\text{Cu K}\alpha$ radiation on a Philips diffractometer. These showed that all the compounds are single-phase materials having the hexagonal CeCo_3B_2 type structure. The lattice parameters obtained are $a = 5.477 \text{ \AA}$ and $c = 3.091 \text{ \AA}$ for CeRh_3B_2 and $a = 5.534 \text{ \AA}$ and $c = 2.999 \text{ \AA}$ for CeRu_3B_2 , which are in good agreement with those reported in the literature (Ku *et al* 1980).

A routine check showed that the CeRh_3B_2 compound, but not the CeRu_3B_2 compound, gets attracted to a magnet at 77 K. This was highly unexpected, particularly since no magnetic ordering has been reported by Ku *et al* (1980) in CeRh_3B_2 . Therefore we carried out careful magnetisation studies in the temperature range 77 to 500 K using the Faraday method. These measurements were made on a commercially supplied susceptibility apparatus with controllable magnetic field and field gradient. The temperature could also be controlled as desired. The results of magnetisation measurements on CeRh_3B_2 and CeRu_3B_2 are shown in figures 1 and 2. The magnetisation of CeRh_3B_2 is fairly large at 77 K even in low applied fields. On heating the sample, a large drop in magnetisation is observed around 115 K which is characteristic of a ferromagnetic to paramagnetic transition. Figure 1 also shows the magnetisation as a function of applied field at 77 K and 300 K. The M - H curve is non-linear and magnetisation seems to show a saturation trend at 77 K, while the M - H curve is a straight line at 300 K, where the system is a paramagnet. In contrast to the behaviour of CeRh_3B_2 , the compound CeRu_3B_2 is a Pauli paramagnet (figure 2) in the temperature range investigated, namely 77–500 K. The cerium ions are most likely quadrivalent (as evidenced also by the unit cell volume) and hence non-magnetic.

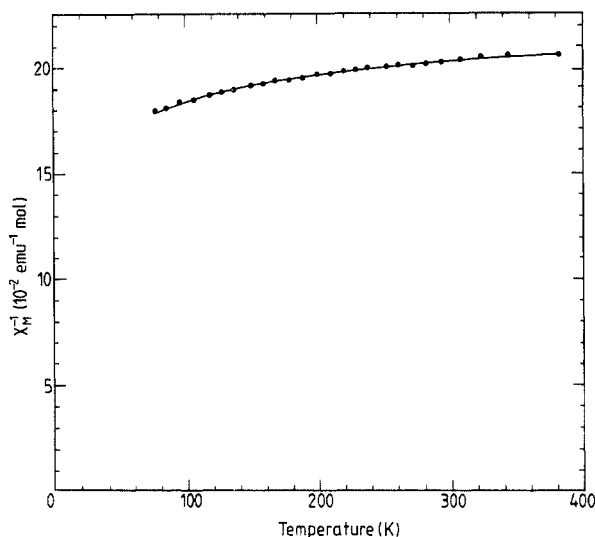


Figure 2. Temperature dependence of inverse molar susceptibility of CeRu_3B_2 .

The magnetic moment per formula unit of CeRh_3B_2 at 77 K and in an applied field of 8 kOe is $0.31\mu_B$. In the paramagnetic state, but well above 115 K, the susceptibility follows a Curie–Weiss behaviour with $\mu_{\text{eff}} = 3.0 \mu_B$ and $\theta_p = -373$ K. The unusually large value of θ_p is most probably associated with the non-integral valence state of cerium ions. The ratio of ordered state moment to the paramagnetic state moment is low, suggesting that the magnetism is of the itinerant type (Moriya 1979) and presumably arises from the Rh d-band. We have also examined the magnetisation of LaRh_3B_2 , PrRh_3B_2 and NdRh_3B_2 prepared by using the same common starting materials. These are found not to order magnetically down to 77 K, although normally Pr and Nd compounds are more paramagnetic than Ce compounds. Ku *et al* (1980) report ordering temperatures of 1.68 K for PrRh_3B_2 and 13.5 K for NdRh_3B_2 but none for the cerium-containing compound. One possibility is that these authors might have missed the magnetic ordering in CeRh_3B_2 due to its low ordered state moment which might escape detection by AC susceptibility measurements. The cerium compound differs from other RRh_3B_2 compounds due to the abnormal valency of cerium ions, and seems to show intrinsic magnetic ordering without magnetic impurities. The promotion of f electrons to the conduction band may also play an important role in inducing magnetic ordering in this compound. In any case, it is interesting that such strong itinerant ferromagnetism is observed in CeRh_3B_2 but not in CeRu_3B_2 or in a few other RRh_3B_2 compounds. We have investigated the effect of a small amount of Fe impurities on the magnetic behaviour of these ternary borides. Preliminary analysis shows that with 99% pure boron, which has typically about 0.5 at. % Fe, the Curie temperature of CeRh_3B_2 is not altered, while the magnetisation increases only marginally. The Pr and Nd compounds with this boron still do not order magnetically down to 77 K.

As mentioned at the beginning, there is an anomaly in the unit cell volume of CeRh_3B_2 which indicates that cerium probably has a valency between +3 and +4 in this compound. The studies on mixed-valent compounds have also attracted a great deal of attention. It is observed that compounds where the rare earth shows mixed-valent behaviour do not, as a rule, order magnetically. We are presently carrying out XPS, x-ray

absorption edge and NMR studies on CeRh_3B_2 to elucidate the valence state of cerium ions both above and below the magnetic ordering temperature. It may also be remarked that during the course of investigating RRh_3B_2 and other borides, we have been able to prepare compounds of the type CeRh_3B_x , where x typically ranges from 0 to 1. These compounds have the AuCu_3 type structure: the same structure as that of CeRh_3 . However, addition of boron results in expansion of the lattice. Since cerium is known to be quadrivalent in CeRh_3 , expansion of the lattice with addition of boron may tend to make cerium trivalent or cause mixed-valent behaviour. Preliminary studies show that CeRh_3B_1 is non-magnetic down to 77 K. Detailed results will be reported elsewhere.

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