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## Effects of carbon on the magnetism of $\text{EuB}_6$

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**Résumé.** — Des mesures d'aimantation ont été effectuées en fonction de la température et du champ magnétique pour divers échantillons  $\text{EuB}_{6-x}\text{C}_x$ . Pour  $x = 0$  et  $x = 0,21$  les composés sont respectivement ferromagnétique et antiferromagnétique. Pour des compositions intermédiaires, ils sont mictomagnétiques.

**Abstract.** — Magnetization has been measured as function of temperature and magnetic field for compounds of composition  $\text{EuB}_{6-x}\text{C}_x$  ( $0 \leq x \leq 0.21$ ). It is suggested that samples with  $x = 0$  and  $0.21$  are ferromagnetic and antiferromagnetic, respectively, whereas those with intermediate composition are mictomagnets.

In a recent paper on the influence of carbon donors on the physical properties of europium hexaboride [1] it was reported that the dominant magnetic interaction in  $\text{EuB}_{6-x}\text{C}_x$  changes from ferromagnetic to antiferromagnetic with increasing carbon content; the paramagnetic Curie temperature decreased progressively from 15 to  $-6.5$  K as  $x$  increased from 0 to 0.21. The  $\text{EuB}_{6-x}\text{C}_x$  system, in contrast with  $\text{Eu}_{1-x}\text{R}_x\text{B}_6$  ( $\text{R} = \text{La}, \text{Gd}, \text{Yb}$ ), has no disorder in the cation sublattice and is therefore an ideal material in which to study the influence of conduction electrons on magnetism. In view of this we decided to obtain the magnetic ordering temperatures  $T_0$  and attempt to establish the magnetic phase diagram of the system, by measuring magnetization as function of temperature and magnetic field for  $x = 0, 0.05, 0.054, 0.082, 0.139$  and  $0.21$ . The purpose of the present paper is to report the results of these observations.

The sintered samples used in these experiments were prepared by borothermal reduction [1]. Magnetization curves at 4.2 K under magnetic fields up to 150 kOe are summarized in figure 1 (<sup>1</sup>). In the figure we have taken into account demagnetization field of  $\frac{4}{3} \pi M$  (3.7 kOe for  $7 \mu_B/\text{Eu}$  ion), assuming that each particle in the powder has a spherical form. Strictly speaking, the demagnetizing factor may vary from 0 to  $4 \pi$ .

We discuss first the magnetization curve for  $x = 0$ . It saturates readily and gradual increase of magnetization at low field may be due to the spread of demagnetizing field and also to the increase of magnetic moment from its thermal average value at 4.2 K

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(<sup>1</sup>) This data was obtained at the Service National des Champs Intenses, Grenoble.

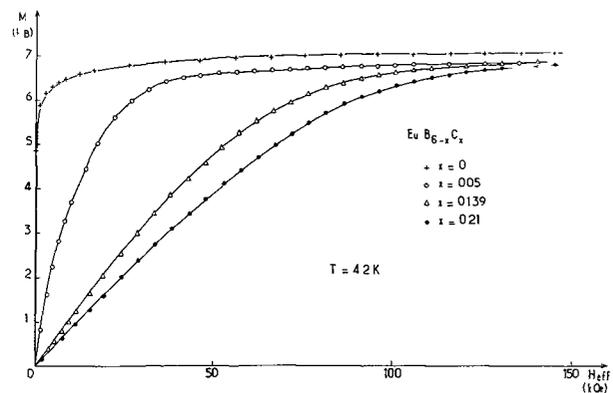


Fig. 1. — Variation of the magnetization as the function of field.

to  $7 \mu_B$ . The curve is therefore characteristic of a ferromagnet whose Curie temperature  $T_C$  is found to be 12.0 K and 12.5 K from Arrott plots and Mössbauer effect [2], respectively. In contrast, for  $x = 0.21$ , the magnetization increases linearly with increasing magnetic field up to 20 kOe, above which it approaches gradually its saturation value. In the present system all the Eu ions are in the  $\text{Eu}^{2+}$  ( $L = 0$ ) state [2] and the anisotropy energy  $H_A$  should be small compared to the exchange field  $H_E$ . In such a case, the magnetization of an antiferromagnetic powder increases nearly linearly with increasing magnetic field at low field and there is no abrupt change from antiferromagnetic to ferromagnetic spin alignment. The magnetization curve for  $x = 0.21$  may be consistent with this scheme. A negative value of the paramagnetic Curie temperature,  $-6.5$  K, also suggests that the  $x = 0.21$  sample is antiferromagnetic. Temperature dependence of the magnetization for  $x = 0.21$  at

1.2 kOe is included in figure 2. In fact there is a small peak at 7.3 K which is observable even at 10 kOe. For an antiferromagnet, the susceptibility at  $T_N$  is

$$\chi(T_N) = 1/A, \quad (1)$$

where  $A$  is the molecular field coefficient, given by

$$A = 6 k T_N / N g^2 \mu_B^2 S(S+1). \quad (2)$$

Putting values of  $T_N = 7$  K and  $S = 7/2$  into eq. (1), we obtain  $\chi(T_N)$  of 0.56 emu/mole, which is reasonable compared to the experimental value of 0.47 emu/mole. From these facts we conclude that the sample with  $x = 0.21$  is an antiferromagnet with  $T_N = 7.3$  K.

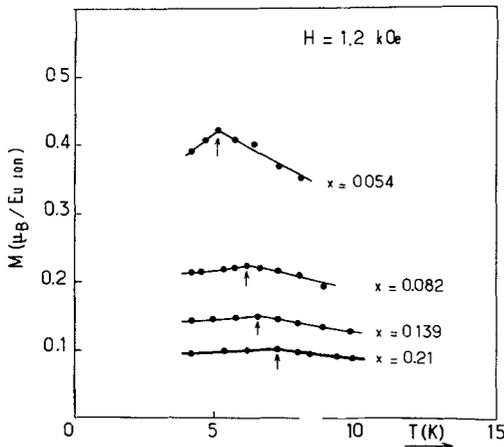


Fig. 2. — Temperature dependence of magnetization of  $\text{EuB}_{6-x}\text{C}_x$  compounds.

Finally we discuss the magnetic properties of  $\text{EuB}_{6-x}\text{C}_x$  for intermediate values of  $x$ . As typical examples, we show magnetization curves at 4.2 K for  $x = 0.05$  and 0.139 in figure 1. The curve for  $x = 0.139$  is essentially same as that for  $x = 0.21$ . However the curve for  $x = 0.05$  is different from those for  $x = 0$  and 0.21. At low fields the susceptibility

is field dependent and saturation of the magnetization is obtained above 50 kOe. Temperature dependences of magnetization at  $H = 1.2$  kOe for  $x = 0.054$ , 0.082 and 0.139 are given in figure 2. There is a peak in the magnetization vs temperature curve. This peak occurs at 5.1 K for  $x = 0.054$  and shifts toward higher temperature with increasing  $x$ . It is worthwhile to note that for  $x = 0.054$  the peak is observable under magnetic fields up to 3.6 kOe, but at higher magnetic fields it disappears. The magnetic ordering temperature for  $x = 0.05$  obtained from Mössbauer effect is 5.2 K [2]. A peculiar feature of the Mössbauer data is the line width for  $x = 0.05$  at 4.2 K which, in contrast to  $x = 0$  or 0.21, is very broad. This suggests that the exchange interactions are inhomogeneous. From these facts there is a strong possibility that the magnetic order of  $\text{EuB}_{6-x}\text{C}_x$  with  $x = 0.05$  is mictomagnetic [3].

Based on the experimental results described above, we show the magnetic phase diagram of  $\text{EuB}_{6-x}\text{C}_x$  system in figure 3. From our experimental results it appears that the zone marked with a query corresponds to a mictomagnetic phase.

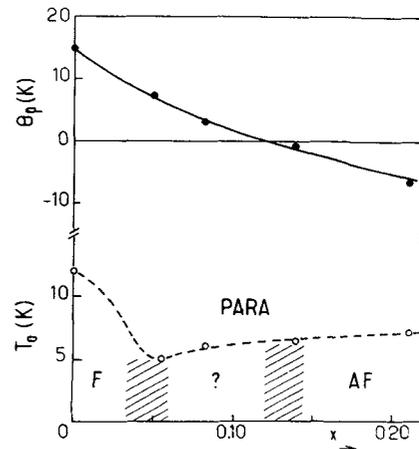


Fig. 3. — Variation of the magnetic ordering temperature and magnetic phase diagram for  $\text{EuB}_{6-x}\text{C}_x$  as a function of  $x$ . For comparison the paramagnetic Curie temperature is also shown in the upper part of the figure.

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