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Magnetic and Electric Properties of EuB_6

By

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The purpose of this note is to report on what, we believe, are the first results of electron spin resonance in a pure single crystal of EuB_6 and to compare them with magnetic and electric data previously published for the same sample /1/. Previous ESR measurements were performed on polycrystalline samples by several authors /2, 3/. Trivalent rare earth ions form hexaborides with metallic behaviour. The same compounds of divalent ions were supposed to be semiconductors. EuB_6 is a cubic material crystallizing in a structure which may be regarded as CsCl-type array of Eu atoms and B_6 octahedra. It has been believed that EuB_6 orders ferromagnetically and contains only Eu^{2+} /4/; but recently Isikawa et al. /5/ have reported magnetic and electric data on this material. Their results indicate that EuB_6 is semi-metallic, contains some Eu^{3+} , and becomes antiferromagnetic at $T_N \approx 6$ K. However, a more recent investigation of carbon substituted EuB_6 /6/ indicates that the paramagnetic Curie temperature decreases smoothly with increasing carbon content, that the order changes from ferromagnetic to antiferromagnetic, and that the Eu is divalent. Kasaya et al. /6/ suggest that the data reported in /5/ may be due to carbon contamination. We felt it would be important to measure the electrical and magnetic properties on the same single crystal of pure EuB_6 . The sample preparation technique has been described previously /1/. ESR measurements were performed at 9 and 35 GHz in the temperature range of 1.4 to 300 K.

In the ESR spectrum, we can clearly separate between two temperature regions below and above ≈ 30 K. In the high temperature range we observed

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Fig. 1. ESR derivate spectrum of EuB_6 single crystal as a function of temperature. $\nu = 9 \text{ GHz}$

one single line with a Dysonian shape which is characteristic of samples with high electric conductivity. A fit with a Lorentzian line shape yields excellent agreement with the observed line shape.

The half linewidth ($\Delta H_{1/2}$), defined as the half-power linewidth of the absorption part of the resonance line can be fitted by the following expression:

$$\Delta H_{1/2} = a + b T,$$

where a at 9 GHz was 750 Oe , at 35 GHz 650 Oe , and $b = (0.3 \pm 0.1) \text{ Oe/K}$ for both frequencies. A value of $g = 1.995 \pm 0.010$ was measured in both cases.

As we decreased the temperature below $\approx 30 \text{ K}$, a clear distortion from a Dysonian line occurred as it is shown in Fig. 1. At the lowest temperature a "resolved" multiple line spectrum was found. The spectrum depended on orientation and strength of the magnetic field as well as on the shape of the sample. Unfortunately we have not been able to separate the different contributions to the Hamiltonian, like dipole-dipole, hyperfine, and crystal field interactions. However, if we compared the ESR data with the electrical resistivity results, shown in Fig. 2, for the same crystal, a minimum in resistivity is clearly seen at $\approx 30 \text{ K}$ (the same temperature at which the ESR line started to distort). This fact can be interpreted as a short range magnetic order.

At 14 K , where our magnetic susceptibility results showed that the material orders ferromagnetically $/1/$, one can see an almost "resolved" spectrum of ESR, together with a drastic minimum in the resistiv-

Fig. 2. Temperature dependence of the electrical resistivity of EuB_6 single crystal

ity that can be understood as a long range ordering.

Hall effect measurements performed for this sample in the range between 77 and 300 K, gave an almost temperature independent negative Hall constant and a net carrier concentration of about 5×10^{19} electrons/cm³. Cohen has performed Mossbauer measurements on our crystals /7/ and found an extremely clean Eu²⁺ spectrum, with less than 0.5% Eu³⁺ possible. To explain the effective moment of $7.76 \mu_B$ per Eu ion obtained from susceptibility, we estimated $\approx 13\%$ Eu vacancy concentration for our single crystals.

The Hall effect indicates a larger contribution of electrons as carriers, whereas band structure considerations /8, 9/ point to hole conduction for Eu vacancies. We concluded that EuB₆ is a semi-metal with some band overlap; where the higher mobility of the electrons related to the mobility of the holes takes account of the negative Hall constant. The ESR, together with the resistivity data, show us that the order is more complicated than one would expect from the susceptibility data alone. Hall experiments as a function of the magnetic field in the range of 1.4 to 77 K are in progress.

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