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## TEMPERATURE DEPENDENCE OF THE FIRST AND SECOND ORDER MAGNETOCRYSTALLINE ANISOTROPY CONSTANTS IN EuS (\*)

G. E. EVERETT and R. A. KETCHAM

Physics Department, University of California, Riverside, California, 92502, U. S. A.

**Résumé.** — Les constantes d'anisotropie du premier et du second ordre,  $K_1$  et  $K_2$ , ont été mesurées en fonction de la température dans le composé ferromagnétique isolant EuS, dont la structure est cfc. Elles ont été déterminées par la variation angulaire de la résonance ferromagnétique à 24 GHz dans des échantillons monocristaux et monodomains. Le modèle à un ion de Wolf n'est pas suffisant pour interpréter nos résultats. Nous pensons pouvoir expliquer ces derniers par la somme des contributions propres des ions et des interactions magnétoélastiques.

**Abstract.** — The first and second order magnetic anisotropy constants,  $K_1$  and  $K_2$ , have been measured in the fcc insulating ferromagnetic compound EuS, as a function of temperature. They have been determined from the angular dependence of the ferromagnetic resonance at 24 GHz in single crystal-single domain samples. The single ion mechanism of Wolf is not sufficient to explain the results. It is suggested that the observed results can be interpreted as the sum of a single ion and magnetoelastic contribution to the anisotropy energy.

The divalent europium chalcogenides are all insulating fcc compounds. Of these, EuO and EuS are ferromagnetic with ordering temperatures of  $T_c = 69$  °K and 16.5 °K respectively. The first order magnetocrystalline anisotropy constant  $K_1^0$  in EuO, has been determined as a function of temperature by Miyata [1] from torque measurements in a (100) plane and the results found to be in good agreement with the single ion theory of Wolf [2]. Hughes [3] has determined the temperature dependence of both the first and second order cubic anisotropy constants from ferromagnetic resonance measurements in a (110) plane. His results for the temperature dependence of  $K_1$  are in good agreement with the results of Miyata [1]. Wolf's theory [2] has been extended by von Molnar [4] to include the higher order cubic crystal field contribution appropriate to the  $S = 7/2$  ground state of the  $\text{Eu}^{2+}$  ion. The result shows poor agreement with the experimentally determined temperature dependence of  $K_2$ .

Von Molnar's [4] calculation considers only the contribution of the cubic crystalline field to the anisotropy energy. Carr [5] has given results for  $\Delta K_1$  and  $\Delta K_2$  arising from inclusion of the second order magnetoelastic constants. From the values of  $\lambda_{100}$  and  $\lambda_{111}$  determined by Argyle [6] one expects  $\Delta K_1/K_1 \sim 1.4 \times 10^{-2}$  in EuO at 4.2 °K. Hughes et al. [3] attribute the deviation of  $K_2$  from the single ion theory as arising from the finite contribution of the second order magnetoelastic coefficients. Using the values of the EuO magnetostriction coefficients for an approximation, we estimate the magnetostrictive contribution to  $K_1$  for EuS should be  $\sim 10\%$  and should be manifest as a deviation from the predictions of the single-ion theory.

**Experimental Procedures.** — The first and second order magnetocrystalline anisotropy constants have been determined from measurements of the angular

dependence of the ferromagnetic resonance field with the external magnetic field being rotated in a (110) plane. The measurements were performed at 23.98 GHz on single crystal-single domain spherical samples, 0.3 mm in diameter prepared from fractionally distilled EuS.

The ferromagnetic resonance was measured using magnetic field modulation and phase sensitive detection. The resulting signal, proportional to the derivative with respect to field of the absorbed power,  $dP/dH$ , goes through zero at the ferromagnetic resonant field and is used in a negative feedback loop to lock the magnetic field to the resonance. This not only improves the precision with which the resonant field could be determined but also made feasible measurements at  $2^\circ$  steps in the interval  $0^\circ$ - $180^\circ$ . For resonance in a (110) plane with  $K_2 \neq 0$ , the angular dependence of the resonant field is expressible in a harmonic series of the form

$$H(\varphi) = H_0 + H_2 \cos 2\varphi + H_4 \cos 4\varphi + H_6 \cos 6\varphi$$

where  $\varphi$  is the angle of the applied field measured from a (100) direction. The data obtained were harmonically analyzed into the various components  $H_0$ ,  $H_2$ ,  $H_4$  and  $H_6$  using a least squares technique. The anisotropy constants expressed as  $(K_1/M)$  and  $(K_2/M)$  were determined from these coefficients. The departure in shape of the sample from a perfect sphere is difficult to measure on this size sample but is less than 1%. Such a small deviation can be regarded as elliptical in its form and thus does not contribute to  $H_4$  or  $H_6$ .

**Results and Discussion.** — The experimentally determined variation with temperature of  $(K_1/M)$  and  $(K_2/M)$  are shown in figures 1 and 2. The  $g$  value was determined to be  $1.970 (\pm 0.001)$  and is temperature independent. We have estimated the absolute uncertainties as being  $\Delta(K_1/M) = \pm 0.5$  gauss and  $\Delta(K_2/M) = \pm 1.6$  gauss. The relative accuracy is determined by the precision with which a particular angle could be reset and is represented by the scatter

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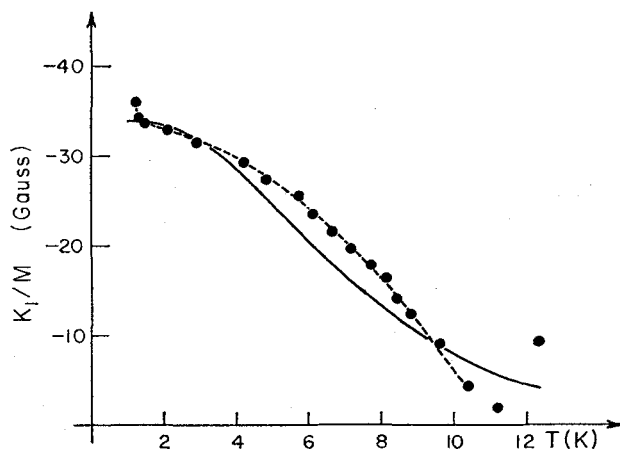


FIG. 1. — The measured first order anisotropy constant,  $K_1/M$ , plotted against temperature. The solid curve is the theoretical dependence obtained by fitting a single ion model to the data at 1.45 °K.

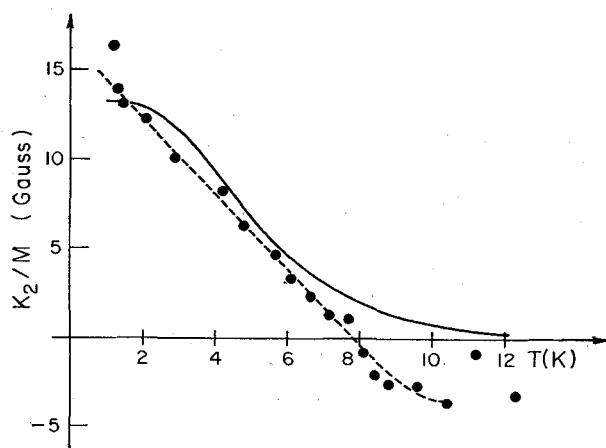


FIG. 2. — The measured second order anisotropy constant,  $K_2/M$ , plotted against temperature. The solid curve is the theoretical dependence obtained by fitting a single ion model to the data at 1.45 °K.

in the data. The points at 12.3 °K should not be considered as significant and are shown only to indicate the effect of a superimposed weak magnetostatic mode on the angular dependence of the resonance field. For temperatures at and below 11.2 °K magnetostatic modes did not interfere with the anisotropy determination.

To compare our results with the single ion theory, the molecular field approximation was used to calculate the temperature variation of the magnetization  $M$  and thus the total effective field. The results for  $K_1/M$  and  $K_2/M$  are shown as solid curves in figures 1 and 2 and correspond to the choice of crystal field parameters  $b_4 = 3.04 \times 10^{-4} \text{ cm}^{-1}/\text{ion}$  and  $b_6 = 0.37 \times 10^{-4} \text{ cm}^{-1}/\text{ion}$ . The parameters correspond to fitting our results at 1.45 °K and serve to illustrate the inadequacy of just the single ion crystal field theory to account for the measured variation with temperature. We also observe that in the range 1.45-8.4 °K, the variation of  $K_1$  is proportional to the seventh power of the reduced magnetization. We conclude that data at lower temperatures would be required to test the tenth power law of  $K_1$ . For the case of  $K_2/M$ , the disagreement is most striking. The fact that  $K_2/M$  changes sign suggests that the anisotropy energy for this material is the result of two mechanisms. The expressions for  $K_1$  and  $K_2$  in terms of the first ( $h_1$  and  $h_2$ ) and second order ( $h_4$  and  $h_5$ ) magnetoelastic coupling constants

$$K_1 = \bar{K}_1 + h_1^2(C_{11} - C_{12}) - 2h_2^2 C_{44}$$

$$K_2 = \bar{K}_2 - 3h_1 h_4(C_{11} - C_{12}) - 12h_2 h_5 C_{44}$$

where  $\bar{K}_1$  and  $\bar{K}_2$  are the anisotropy energies at zero strain and stress. The samples measured are annealed after grinding and mounted with a minimum of adhesive. Even so, we suspect that there is some stress resulting from the differential thermal contraction between the sample and mounting adhesive. If we represent the total anisotropy energy as  $K = \bar{K} + \Delta K$  and assume that  $\bar{K}$  is given by the single ion theory, the resulting variation of  $\Delta K$  for various choices of  $b_4$  and  $b_6$  is not monotonic in the temperature. Also, the rather abrupt increase in both ( $K_1/M$ ) and ( $K_2/M$ ) at the lowest temperature is not understood.

We summarize our results as demonstrating that the single ion mechanism is not sufficient to account for the observed magnetocrystalline anisotropy energy in EuS. They show that there is a second mechanism contributing, which we suggest could be due to magnetostriction. This interpretation requires that the second order magnetoelastic coefficients make a substantial contribution to the magnetoelastic energy in this material.

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