Effect of a Uniaxial Stress on the Curie Temperature of a Single Crystal of Gd

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The variation of the Curie temperature of a single crystal of Gd when subjected to a uniaxial stress up to 130 bar applied along the c axis of the crystal, is $(-1.55\pm0.05)\times10^{-3}$ deg bar^{-1}. The rate of change with a stress applied along the a or b axis is $(0\pm0.05)\times10^{-3}$ deg bar^{-1}. The variations of the exchange interaction with the a, b, and c crystallographic parameters are deduced, together with the variation of the Curie temperature with hydrostatic pressure, the magnetic length anomaly along the various crystallographic axes, and the anisotropic forced magnetostriction. The calculated values are in good agreement with the experimental values.

INTRODUCTION

WE have measured the variation of the Curie temperature of a single crystal of Gd when a uniaxial stress up to 130 bar is applied along the a, b, or c crystallographic axis (Fig. 1). In previous work[1-3] we studied the effect of hydrostatic pressure on the magnetic ordering temperatures of rare-earth single crystals (Gd, Tb, Dy). In order to interpret the experimental results, we used a one-dimensional model in which only the variation of exchange energy with the c parameter of the crystallographic lattice was considered. This model was justified by the fact that the variation of the Curie temperature of polycrystalline gadolinium is higher than the variation of the Curie temperature of a single crystal. This arises because the value of the linear compressibility $K_{\parallel}$ measured parallel to the c axis at 298°K is less than the average compressibility $\frac{1}{3}(K_{\parallel}+2K_{\perp})$.[3] This model allowed for the calculation of the magnetic contribution to the thermal expansion of the c parameter of Gd, Tb, and Dy. The calculated values were in good agreement with those determined experimentally for Dy. The agreement was not as satisfactory for Gd and Tb. Consequently, we thought that it would be necessary to determine the variation of exchange energy along the a, b, and c crystallographic axes. The results of this investigation are reported herein. The Gd sample was a single crystal with a specified purity of 99.9%, purchased from Metals Research, Cambridge. It was cut into a 4-mm cube with the a, b, and c crystallographic axes perpendicular to the cubic faces (Fig. 1) within 2°.

I. APPARATUS

The Curie temperature of Gd was determined from the temperature variation of the susceptibility of the sample placed in a weak alternating field whose frequency was 495 cps.[3] The pressure was generated by placing known weights on the plate (a) (Fig. 2). The total weight is applied to a face of the sample by means of a mechanical system having one degree of freedom. The mechanical part which transmits the force from the plate to the sample is restrained by a ball bearing arrangement (b). The sample (c) is mounted on a ball and socket joint (d). The applied force can be determined to within 1%. The sample is surrounded by a mutual inductance M_0 (e) whose value is proportional to the susceptibility of the sample. The temperature was measured by means of a Cu-constantan thermocouple (f) whose junction is in thermal contact with the Be-Cu sample holder. The experimental cell (g) is insulated from its support (h) with epoxy glass (i). The temperature of the sample can be varied by means of cooling water flowing in the heat exchanger. The whole system is insulated with quartz wool (k).

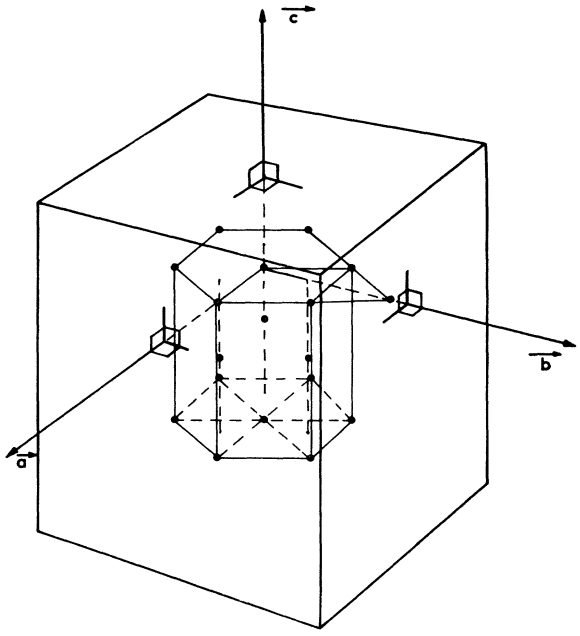

FIG. 1. Crystallographic orientation of the cube used in the experiment.

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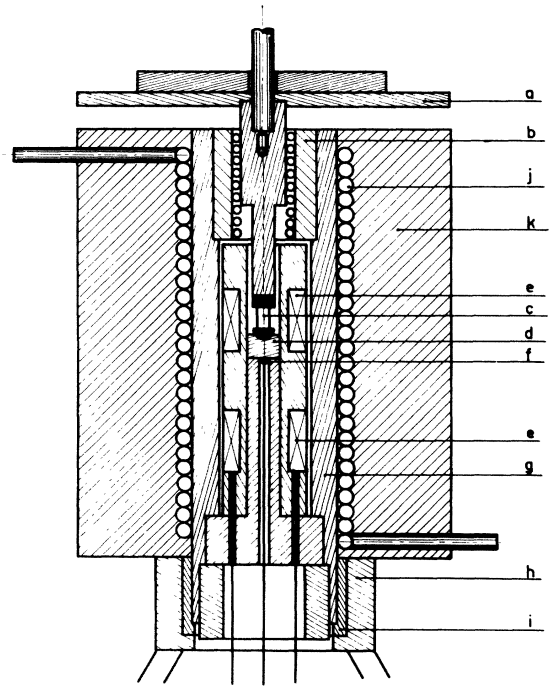

FIG. 2. Apparatus for the application of uniaxial stress and measurement of the Curie temperature: (a) plate; (b) ball-bearing arrangement; (c) sample; (d) ball and socket joint; (e) coils for the measurement of the susceptibility; (f) Cu-constantan thermocouple junction; (g) experimental cell; (h) support of the experimental cell; (i) epoxy glass; (j) heat exchanger; and (k) quartz wool.

II. EXPERIMENTAL RESULTS

The temperature dependence of the Curie point was determined from the temperature dependence of the susceptibility of the sample for a given stress. When the magnetic field generated by the coil and the applied stress are directed along the c axis (Fig. 3), initially with increasing temperature, the susceptibility shows a slight decrease, then remains constant over a range of a few tenths of a degree, and finally decreases around the Curie temperature. Thus, the temperature dependence of the susceptibility can be represented by two straight lines D_1 and D_2. At different pressures, the relative susceptibility curves can be deduced by parallel translations. In order to determine the variation of the Curie temperature as a function of the stress applied along the c axis, we have drawn the straight line D_2' parallel to D_2, which intersects D_1 in the neighborhood of the inflection point of the susceptibility curve A'. We assume that the variation of the temperature associated to A' is equal to the variation of the Curie temperature. When the stress is applied along the a or b axis, no variation of the Curie temperature is observed up to 130 bar (Fig. 4). Thus, we obtain

$$(\partial\theta/\partial p)_c = -(1.55\pm0.05)\times10^{-3}\ \text{deg bar}^{-1},$$
$$(\partial\theta/\partial p)_a = (\partial\theta/\partial p)_b = (0\pm0.05)\times10^{-3}\ \text{deg bar}^{-1}.$$

The experimental value given in Ref. 3 is

$$\partial\theta/\partial p = -(1.40\pm0.02)\times10^{-3}\ \text{deg bar}^{-1}$$

for hydrostatic pressure.

III. VARIATION OF CURIE TEMPERATURE WITH PRESSURE AND VARIATION OF EXCHANGE INTERACTIONS WITH LATTICE PARAMETERS

Within the molecular field model, the Curie temperature θ of Gd is given by the relation[4]

$$k\theta = \tfrac{2}{3}S(S+1)I, \qquad (1)$$

where $S=\frac{7}{2}$. I is the exchange interaction between a magnetic ion and the neighboring ions; it can be developed as a function of the variations e_{aa}, e_{bb}, and e_{cc} of the lattice parameters. As a first-order approximation, we write

$$I(a,b,c) = I(a_0,b_0,c_0) + (\partial I/\partial a)_0 a_0 e_{aa} + (\partial I/\partial b)_0 b_0 e_{bb} + (\partial I/\partial c)_0 c_0 e_{cc}. \qquad (2)$$

Then we have

$$k\theta = \tfrac{2}{3}S(S+1)[I(a_0,b_0,c_0) + (\partial I/\partial a)_0 a_0 e_{aa} + (\partial I/\partial b)_0 b_0 e_{bb} + (\partial I/\partial c)_0 c_0 e_{cc}]. \qquad (3)$$

The variation of the Curie temperature with stress can therefore be written as a function of the variations of the lattice parameters:

$$\partial\theta/\partial p = \tfrac{2}{3}S(S+1)[T_1(1/a_0)\partial a/\partial p + T_2(1/b_0)\partial b/\partial p + T_3(1/c_0)\partial c/\partial p], \qquad (4)$$

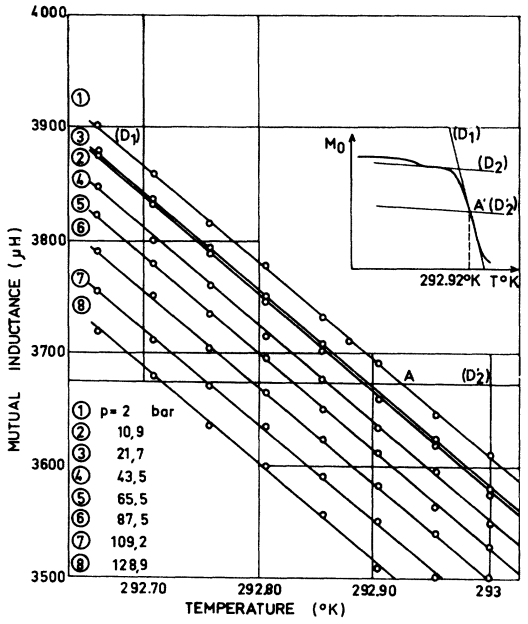

FIG. 3. Thermal variation of the mutual inductance of the measuring coils—stress and magnetic field are applied along the c axis.

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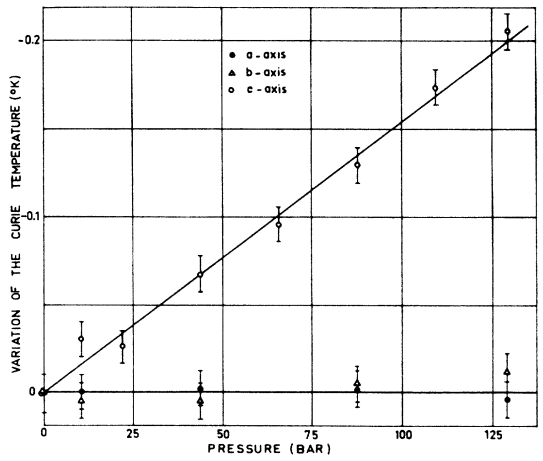

FIG. 4. Variation with uniaxial stress of the Curie temperature of a single crystal of Gd.

with

$$T_1=\left(\frac{\partial(I/k)}{\partial \ln a}\right)_0;\quad T_2=\left(\frac{\partial(I/k)}{\partial \ln b}\right)_0;\quad T_3=\left(\frac{\partial(I/k)}{\partial \ln c}\right)_0.$$

From the experimental results and the values of the elastic moduli,[5] we obtain

$$\begin{aligned}&10.5\times10^{-6}(0.3757T_1+0.3757T_2\\&\qquad-1.614T_3)=-1.55\times10^{-3},\\&-1.833T_1+0.566T_2+0.3757T_3=0,\\&0.566T_1-1.833T_2+0.3757T_3=0.\\&10.5\times10^{-6}(-0.8917T_1+0.8917T_2\\&\qquad-0.8624T_3)=-1.40\times10^{-3}.\end{aligned}\tag{5}$$

We have taken, as a first approximation, the isothermal elastic moduli equal to the adiabatic coefficients. Then, we have

$$T_1=T_2=25°\mathrm{K};\quad T_3=103°\mathrm{K}.\tag{6}$$

These values can be compared to those previously obtained with the one-dimensional model[3]:

$$T_1=T_2=0°\mathrm{K};\quad T_3=\frac{2d(A_1/k)}{d\ln c}=150°\mathrm{K}.$$

IV. MAGNETIC LENGTH ANOMALIES

The magnetocrystalline anisotropy of Gd is weak. Thus the free energy per unit volume, G, can be written[3,6]

$$G=\tfrac{1}{2}C_{33}e_{ee}^2+2C_{13}e_{aa}e_{cc}+(C_{11}+C_{12})e_{aa}^2+\bar{N}IM^2S^2-\bar{N}kT\ln\left[\sum_{m=-S}^{+S}\exp\left(\frac{m}{kT}(g\mu_BH+2ISM)\right)\right].\tag{7}$$

C_{ij} designates the elastic moduli[5] and M the relative magnetization. The relation (7) is justified from the experimental point of view since one cannot separate the magnetic length anomalies e_{aa} and e_{bb}. In expression (7) the elastic and the magnetic energies have been taken into account. By minimizing G, one obtains

$$\begin{aligned}e_{aa}&=\frac{k\bar{N}S^2M^2}{C_{33}(C_{11}+C_{12})-2C_{13}^2}(C_{33}T_1-C_{13}T_3),\\e_{cc}&=\frac{k\bar{N}S^2M^2}{C_{33}(C_{11}+C_{12})-2C_{13}^2}[(C_{11}+C_{12})T_3-2C_{13}T_1].\end{aligned}\tag{8}$$

The thermal variation of the magnetic length anomaly along the a and c axes has been calculated from the values of T_1 and T_2 and the elastic moduli has been determined between 4 and 300°K [Eq. (5) (and Fig. 5)]. The thermal variation of the magnetization has been calculated from the Brillouin function $B_J(Z)$ relative to the quantum number J. The experimental curves for the thermal variation of e_{aa} and e_{cc} were deduced from the measurements of Bozorth and Wakiyama[7]; these curves have been corrected for the thermal dilatation of the nonmagnetic lattice. This contribution has been determined by a linear extrapolation down to 100°K of the nonmagnetic high-temperature curve.[7,8] This extrapolation is justified by the low value of the Debye

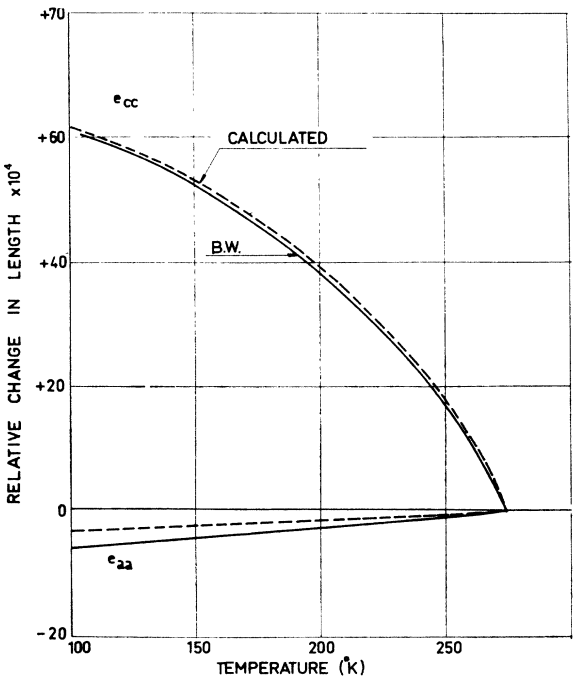

FIG. 5. Thermal variation of the magnetic length anomalies along the c and a axes of a Gd single crystal calculated from values from (8). Experimental values from Bozorth and Wakiyama (B.W.) corrected for the thermal dilatation of the nonmagnetic lattice.

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temperature of Gd (approximatively 180°K).[9,10] The calculated values for e_{aa} and e_{cc} are in good agreement with experimental ones. The magnetic length anomaly along the c axis, when extrapolated to 0°K, is equal to 6.5×10^{-3}, compared to 3×10^{-3} obtained by Darnell[7] and Ergin[10] and to 6.5×10^{-3} deduced from the experimental results of Bozorth and Wakiyama.[7]

V. ANISOTROPIC FORCED MAGNETOSTRICTION

One can use the relationships (8) in order to determine the anisotropic magnetostriction coefficients along the a ($\lambda_\perp$) and c (λ_{11}) crystallographic axes. The relative magnetization M is a function of the applied magnetic field:

$$M=B_j(Z)$$

with

$$Z=\frac{g\mu_B SH}{kT}+\frac{3SB_j(Z)\Theta}{(S+1)T}, \tag{9}$$

$$\lambda_\perp=\frac{2\bar{N}g\mu_B S^3}{\Theta}\frac{[(C_{33})T_1-C_{13}T_3]}{C_{33}(C_{11}+C_{12})-2C_{13}^2}\times\frac{MM'}{(T/\Theta)-3SM'/S+1},$$

$$\lambda_{11}=\frac{2\bar{N}g\mu_B S^3}{\Theta}\frac{[(C_{11}+C_{12})T_3-2C_{13}T_1]}{C_{33}(C_{11}+C_{12})-2C_{13}^2}\times\frac{MM'}{(T/\Theta)-3SM'/S+1}, \tag{10}$$

where M' is the derivative of the relative magnetization with respect to Z. The value $\lambda_{11}/\lambda_\perp$ is a function of the elastic moduli and of the coefficients T_i. It is practically independent of the temperature, a result which is in good agreement with the experiment described in Ref. 7. In Fig. 6, the experimental results by Bozorth and Wakiyama[7] for magnetic fields between 10 and 20 kOe, are given, together with the experimental results of Coleman and Pavlovic[12] for magnetic fields between 11 and 14 kOe. The calculated values of λ_{11} and $\lambda_\perp$ (Fig. 6) have been determined from Eq. (10) for 15 kOe. It can be seen that the forced magnetostriction coefficients are approximatively independent of the value of the applied field (9), which is in good agreement with the experiments mentioned above. Figure 6 shows that there is satisfactory agreement between the experimental and the calculated values of the forced magnetostriction coefficients. The use of a one-dimensional model[3] which considers only the variation of exchange

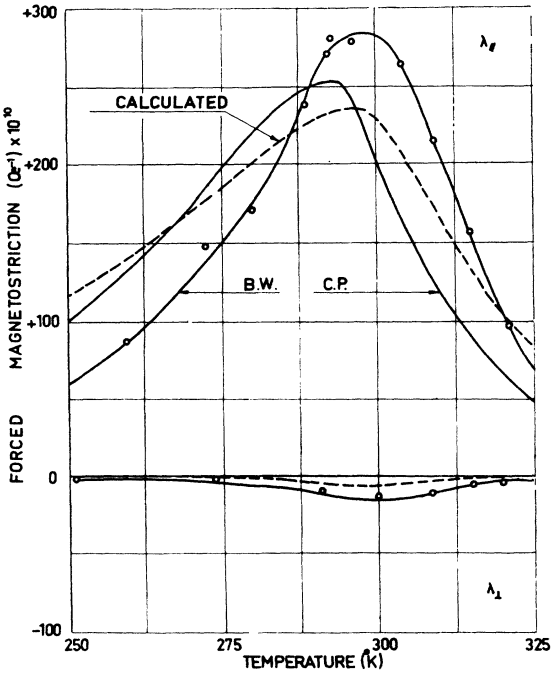

FIG. 6. Thermal variation of the forced magnetostriction along the c and a axes of a Gd single crystal calculated from values from (10). Experimental values from Bozorth and Wakiyama (B. W.) and Coleman and Pavlovic (C. P.).

interactions with the c parameter is a good approximation for the determination of magnetic length anomaly and forced magnetostriction along the c axis. In fact, the relationship giving these quantities (8) and (10), are characterized by a term,

$$2C_{13}T_1/(C_{11}+C_{12})T_3,$$

which is ~0.12 and is thus practically negligible compared to 1. Relationships (8) and (10) show that the one-dimensional model is not satisfactory for the determination of the magnetic length anomaly and the forced magnetostriction coefficients along directions which are perpendicular to the c axis.

VI. CONCLUSION

The analysis of our experimental results shows that the study of the effect of uniaxial stress on the magnetic ordering temperature of a single crystal constitutes a new and useful technique. It allows the interpretation of the effect of hydrostatic pressure on magnetic ordering temperatures, as well as the interpretation of spontaneous and forced anisotropic magnetostriction. Magnetostriction phenomena play an important role in the magnetic properties of rare-earth metals. Studies of the variation of magnetic ordering temperatures, magnetization, threshold field, and magnetocrystalline anisotropy of rare-earth metals with hydrostatic pressure and uniaxial stress will allow a better understanding of their magnetic properties.

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