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Magnetic Anisotropy of Gd Metal at 4 K under Pressure

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Measurements are presented of the magnetic crystalline anisotropy of a spherical single crystal of gadolinium metal of high purity (about 0.01 wt.% oxide) at 4.2 K, at atmospheric pressure and at near-hydrostatic pressures up to 6 kbar. The magnetic anisotropy is strongly pressure dependent. It is suggested that part of this anisotropy is caused by the movement through the Fermi level of a small peak in the density of electron states curve.

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The magnetic properties of gadolinium metal are receiving renewed interest since single crystals of highly purified, oxygen-free material became available. Smith, Tanner, and Corner¹ showed that the large divergence in the magnetic anisotropy data for gadolinium metal is essentially due to the presence of gadolinium-oxide platelets perpendicular to the c axis. An oxide content of not more than 1 or 2 wt.% may cause changes in the first anisotropy constant of 100% or more, depending on temperature. In particular the discrepancy between neutron-diffraction and magnetic-torque data on the direction of the easy axis of magnetization could be resolved by their analysis.

The magnetic anisotropy of Gd is small compared to that of the other rare-earth metals. This is in accordance with the fact that the orbital moment of the half-filled $4f$ shell is zero. In first order there is no spin-orbit coupling for the localized $4f$ electrons. Part of the magnetic moment, however, arises from polarization of the nonlocalized $5d$ electrons. We conclude from our experiments that these electrons are mainly responsible for the anisotropy.

The magnetic torque of a gadolinium single-crystal sphere has been measured at temperatures from 4.2 to 300 K and at pressures from 1 bar up to 6 kbar. Only the 4.2-K measurements will be presented in this Letter. We used one of the single-crystal spheres on which Roeland *et al.*² performed high-field magnetization measurements (sample B₁, diameter 2.8 mm, mass 0.0911 g). The sphere was spark cut and annealed afterwards at 900 °C during 24 h. The residual resistivity ratio was about 150, the oxide content about 0.01 wt.%. The sample orientation was determined within 2° by x-ray techniques.

Torque measurements were carried out with the field (1.8 T) in the (c, a) plane in a conventional torque magnetometer.³ The directions of the c and a axes could be determined within 0.1° from

the measured torque curves. The specimen was mounted in a small beryllium-copper pressure cell, connected with the pressure-generating unit by a thin (1-mm diam) capillary tube. In order to achieve hydrostatic conditions at 4.2 K, helium gas was frozen at constant pressure. Pressures were read at this temperature by means of strain gauges mounted on the pressure cell.

Some experimental torque curves at 4.2 K are shown in Fig. 1. The torque L_A in this figure is given as a function of the angle θ between the direction of the magnetization and the c axis (the difference of directions of magnetization and external field has been corrected for). The curves show a complicated θ dependence and Fourier coefficients at least up to degree 24 have to be evaluated before a fair representation of the data is

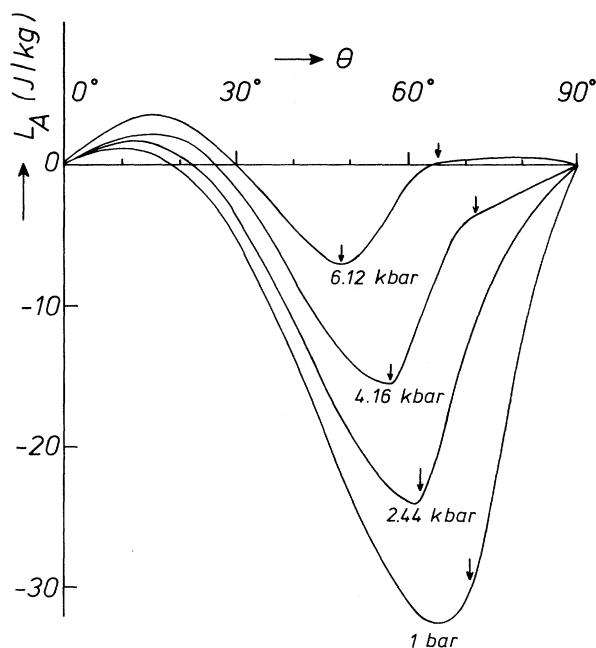


FIG. 1. The magnetic torque of gadolinium in the (c, a) plane at 4.2 K at different pressures. The edges of the "transition region" are marked by small arrows.

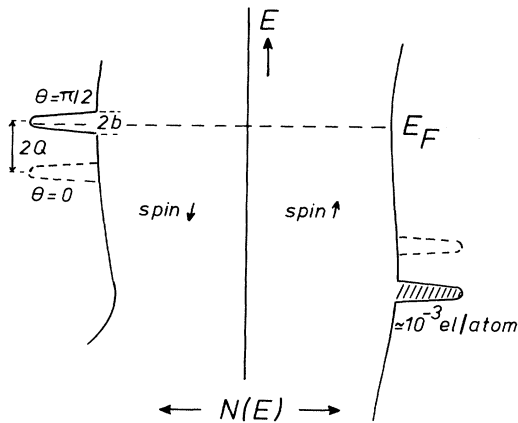


FIG. 2. Schematic representation of the density of states for minority- and majority-spin electrons in accordance with the proposed model.

reached. We therefore suggest here another analysis, related to the one that has been applied earlier to Ni metal at 4.2 K.⁴

We assume that the anisotropy energy consists of two parts: a regular part that contains only a few Fourier components of low degree and that can therefore be described with a few anisotropy constants, and an anomalous part that is connected with the movement through the Fermi level of a peak in the density-of-states curve.

The model we are proposing for the anomalous part is schematically shown in Fig. 2. A peak in the density-of-states curve, due to a flat part in one particular energy band, is situated close to the Fermi energy for the minority-spin electrons. This peak can shift through the Fermi level with a variation of pressure (p), magnetic field (B), or the angle of the direction of the magnetization with the c axis (θ). From an experimental point of view we cannot distinguish which of the two subbands (the minority- or the majority-spin electron subband) is responsible for the anomaly observed in our magnetic torque measurements. Upon rotation of the magnetization the two peaks in the density-of-states curves will shift in opposite directions as is indicated in Fig. 2. As long as both peaks remain below the Fermi level the contributions to the anisotropy energy from these particular energy states cancel each other. If the peak of the minority-spin electron subband is well above the Fermi level we may expect contributions to the anisotropy energy that are proportional to the θ -dependent energy shift and the number of unoccupied energy states in this peak.

In accordance with the uniaxial crystal sym-

metry we write in first order for the difference in energy between the center of the peak and the Fermi level

$$\epsilon = -Q \{ \cos 2\theta - C(p) \} + \mu_B (B - B_0), \quad (1)$$

where Q is a parameter, proportional to the spin-orbit interaction. The parameter $C(p)$ is approximated by $C(p) = C_0 + \lambda p$, which means that the shift in energy of the center of the peak with pressure is given by λQ .

Of course, only the sum of the two constant terms in Eq. (1) ($QC_0 - \mu_B B_0$) is significant. We use this notation since not all parameters in Eq. (1) can be derived from measurements at one single value of B . If we choose B_0 equal to the magnetic field value actually used, the second term in Eq. (1) can be dropped and $C(p)$ can be determined accurately even if Q can not.

The width of the peak, which will be assumed to have a value $2b$, is not small compared to its shift and we have to deal with a transition region in which the Fermi level shifts through the peak. Denoting the number of states in this peak by n we can write for the anomalous contribution to the anisotropy energy

$$\begin{aligned} E_A' &= n\epsilon \text{ for } \epsilon > b, \\ E_A' &= nb f(\epsilon/b) \text{ for } -b < \epsilon < b, \\ E_A' &= 0 \text{ for } \epsilon < -b. \end{aligned} \quad (2)$$

Here, $f(x)$ (defined for $-1 < x < 1$) is a smooth function of x , approximated by $K(1+x)^{5/2}$ for x near -1 , and by $x + K(1-x)^{5/2}$ for x near $+1$. These $5/2$ powers result from the assumption of a quadratic dependence of ϵ on the wave vector k near the top and bottom of the flat part of the energy band of interest. In our analysis we used

$$\begin{aligned} f(x) &= (2/\pi) \int_{-1}^x (x - \epsilon)(1 - \epsilon^2)^{1/2} d\epsilon \\ &= \frac{1}{2}x + (2/3\pi)(1 + \frac{1}{2}x^2)(1 - x^2)^{1/2} \\ &\quad + (x/\pi) \arcsin x. \end{aligned} \quad (3)$$

The results of our analysis are not very sensitive to the exact shape of $f(x)$.

For the magnetic torque we derive now the following expression:

$$L_A = -\frac{\partial E_A}{\partial \theta} = \sum_{i=1}^4 S_{2i} \sin(2i\theta) - \frac{\partial E_A'}{\partial \theta}, \quad (4)$$

where the coefficients S_{2i} represent the regular part of the magnetic torque. Note more than four coefficients, S_2 , S_4 , S_6 , and S_8 , are needed to describe this regular part of the anisotropy of Gd at 4.2 K within the experimental accuracy. Val-

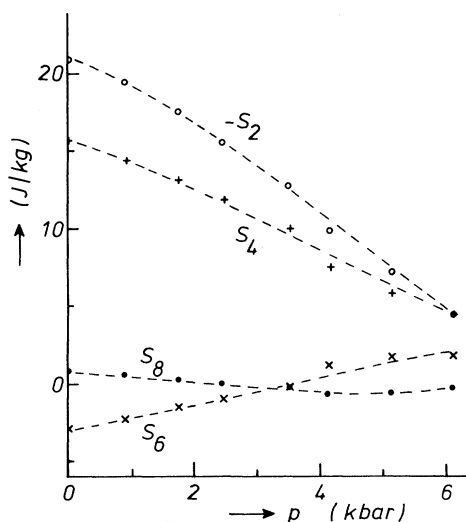


FIG. 3. The Fourier coefficients S_2 , S_4 , S_6 , and S_8 , representing the regular part of the anisotropy of Gd at 4.2 K, for different pressures up to 6.10 kbar.

ues for these coefficients are shown in Fig. 3. Equation (4) can be rewritten as

$$\begin{aligned}
 L_A &= \sum_{i=1}^4 S_{2i} \sin(2i\theta) \text{ for } \epsilon < -b, \\
 L_A &= \sum_{i=1}^4 S_{2i} \sin(2i\theta) \\
 &\quad + 2nQf'(\epsilon/b) \sin 2\theta \text{ for } -b < \epsilon < b, \\
 L_A &= \sum_{i=1}^4 S_{2i} \sin(2i\theta) + 2nQ \sin 2\theta \text{ for } \epsilon > b.
 \end{aligned} \tag{5}$$

In these expressions ϵ is given by Eq. (1) with the second term dropped. If all three regions for ϵ occur, a fit of the measured torque curve at a given pressure p yields values of S_2 , S_4 , S_6 , S_8 , nQ , $C(p)$, and b/Q .

In Gd metal at pressures below 2 kbar only the first and second regions were found to be present. A unique determination of the parameters nQ , $C(p)$, and b/Q is impossible in that case. An analysis of the torque data of Gd above 2 kbar, however, results in values for b/Q that are essentially independent of pressure; see Fig. 4. Taking a constant value of 0.23 for b/Q , we are able to derive values for $C(p)$ below 2 kbar. The $C(p)$ values calculated in this way satisfy the relation $C(p) = C_0 + \lambda p$ with the same values for C_0 and λ as obtained from the experiments above 2 kbar ($C_0 = -1.023$ and $\lambda = 0.10 \text{ kbar}^{-1}$).

A point of further discussion in this interpretation of the magnetic torque data of Gd remains the strong pressure dependence shown by the pa-

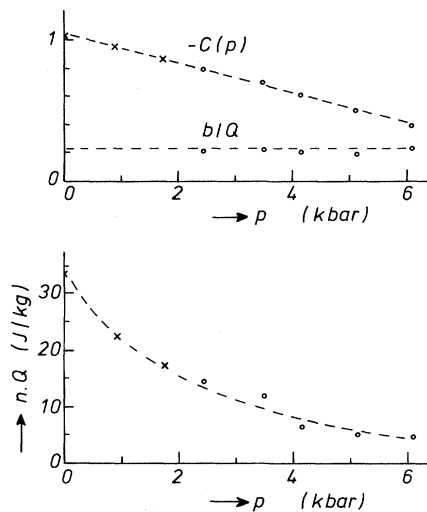


FIG. 4. The parameters $C(p)$, b/Q , and nQ describing the anomalous part of the anisotropy energy of gadolinium at 4.2 K. Values for $C(p)$ and nQ below 2 kbar have been calculated assuming a value for b/Q of 0.23, see text.

rameter nQ . This parameter changes from 33.8 J/kg at a pressure of 1 bar to 4.7 J/kg at a pressure of 6.10 kbar. This pressure dependence is not yet understood. Large pressure effects, however, are also observed in the regular part of the anisotropy energy, i.e., the parameters S_{2i} , see Fig. 3. A strong influence of pressure on cross sections of the Fermi surface of Gd has also been reported by Schirber *et al.*⁵

An additional experiment at 25 K and 6 kbar gave nearly the same values for all parameters as those obtained at 4.2 K and 6.19 kbar, except for the parameter b/Q . This latter parameter has at 25 K about twice its low-temperature value. If we assume this extra broadening to be due to a temperature effect and therefore proportional to kT , we roughly estimate that $Q \cong 14 \text{ meV}$. It then follows that n , the number of states contained in the peak, is about $5 \times 10^{-4} e/\text{at}$. Both values belong to a pressure of 6 kbar; at lower pressures, n (or Q) is considerably larger.

This value for n is too small to expect a readily detectable change in the magnetic moment of Gd at the peak crossing the Fermi level. A preliminary experiment on the pressure dependence of the magnetic moment of Gd, measured along the b axis, resulted in a value of $(0 \pm 1) \times 10^{-3} \mu_B/\text{kbar}$ over the pressure range 0–4 kbar. Experiments are planned in which this anomaly will be further investigated as a function of pressure, tempera-

ture, and magnetic field intensity.

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Charge Transport in the Organic Conductor Quinolinium Ditracyanoquinodimethanide [Qn(TCNO)₂]

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Proton spin-lattice relaxation-time (T_1) measurements are reported in pure and in neutron-irradiated organic conductor Qn(TCNO)₂. The diffusion constant D (evaluated from T_1) and conductivity σ have the same dependence on the irradiation-induced impurity concentration, showing that charge propagation is responsible for the relaxation time. The temperature dependence of D , when compared with $\sigma(T)$, suggests that the conductivity is determined by the temperature-dependent mobility.

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In spite of considerable effort, charge transport in organic, highly anisotropic conductors is only poorly understood, and various models have been proposed to account for the temperature dependence of the conductivity $\sigma(T)$ in various materials. Qn(TCNO)₂ is a member of the TCNQ salts,¹ which show metalliclike conductivity at high temperatures, followed by a broad maximum and decreasing σ with decreasing temperatures. This behavior was suggested to be due to a smeared metal-to-insulator transition,² to a small-band-gap semiconductor type of behavior³ where $\sigma = \mu ne$ with the number of carriers n increasing exponentially with temperature and μ strongly decreasing with increasing T . A model based on the effect of disorder⁴ accounts for $\sigma(T)$ through a temperature-dependent mobility.

In this paper we discuss proton NMR relaxation experiments, T_1 , in pure and also in neutron-irradiated Qn(TCNO)₂. We argue that T_1 is determined by the charge diffusion, and is thus directly related to the conductivity. We also show that σ and the diffusion constant have a similar tem-

perature dependence, strongly suggesting that the carrier concentration is only weakly dependent on temperature.

The proton spin-lattice relaxation rate is given by⁵

$$T_1^{-1} = \Omega^z F(\omega_N) + \Omega^+ F(\omega_e), \quad (1)$$

$$F(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \langle s^z(t) s^z(0) \rangle,$$

where $\Omega^z = \frac{3}{5} d^2$ and $\Omega^+ = a^2 + \frac{7}{5} d^2$ are coupling constants, reflecting hyperfine, a , and dipolar, d , electron-nuclear coupling; ω_N and ω_e are the nuclear and electronic frequencies. The spin correlation function $\langle s^z(t) s^z(0) \rangle$ depends on the dimensionality of the diffusion. For one-dimensional (1D) diffusion, $\langle s^z(t) s^z(0) \rangle \sim (Dt)^{-1/2}$, where D is the diffusion constant, while for 3D diffusion $\langle s^z(t) s^z(0) \rangle \approx (Dt)^{-3/2}$. This leads, through Eq. (1), to a strongly frequency-dependent T_1 for 1D diffusion,^{5,6} and T_1 nearly independent of ω for a 3D diffusion process. The spin-lattice relaxation rate samples the propagation of spins, times the effective number of spins, and can be rewritten