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Neutron Diffraction Study of the Magnetic Behavior of Gadolinium*

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Neutron diffraction measurements were made on a single crystal of gadolinium at sample temperatures from 10 to 350°K to investigate the possible occurrence of a spiral spin structure. The measurements show that Gd is a normal ferromagnet with a rather complex temperature dependence of the spontaneous moment alignment. Between $T_C=294$ and 232°K the moment is parallel to the c axis; below 232°K it moves away from the c axis to a maximum deviation of about 65° near 180°K and then back to within 32° of the c axis at low temperatures.

INTRODUCTION

GADOLINIUM is ferromagnetic with a Curie temperature of 293°K[1,2] and a saturation moment of 7.55 μ_B/atom.[1] Recently, Belov and Pedko[3] observed low-field magnetization anomalies in polycrystalline Gd above 210°K and concluded that a spiral spin structure similar to that observed in the other heavy rare-earth metals also occurred in Gd, but that small fields (0–15 Oe in the 210–290°K temperature region) were sufficient to transform the spiral into a ferromagnet. Neutron diffraction measurements[4] did not reveal any satellite reflections of the type expected for a spiral spin structure analogous to the other heavy rare earths. However, these measurements were made at a neutron wavelength of 1.055 Å for which the absorption cross section is so large that intensity measurements were limited to those reflections with scattering vectors nearly normal to the face of the crystal. Recently, we obtained a 0.353-Å beam of neutrons from the ORR. At this wavelength the resonance absorption cross section of Gd is smaller by two orders of magnitude and a more complete neutron study can be made.

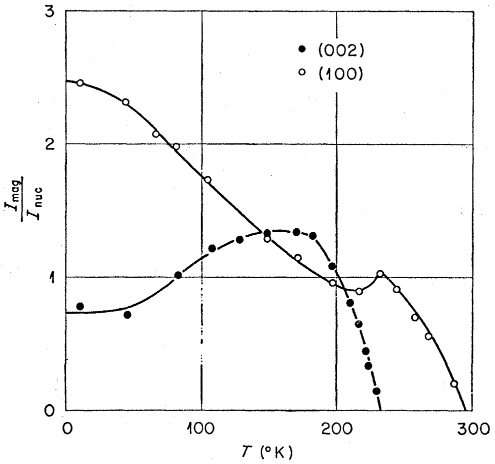

FIG. 1. Temperature dependence of the magnetic to nuclear intensity ratios for the (100) and (002) reflections of Gd.

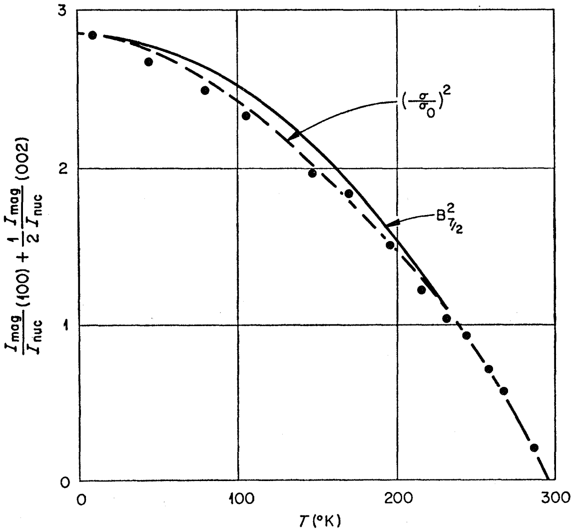

FIG. 2. Temperature dependence of the sum

$$(I_{mag}/I_{nuc})\,(100)+\tfrac{1}{2}(I_{mag}/I_{nuc})\,(002).$$

Also shown are a squared $\frac{7}{2}$ Brillouin function and the square of the relative magnetization (Ref. 1) normalized to the low-temperature data point.

RESULTS

The Gd single crystal was a flat disc 5 mm in diam and 0.5 mm thick with the c axis normal to the face of the disc. Peak-intensity measurements were made for the (100) and (002) reflections as a function of temperature from 10 to 350°K. Calculations based on a Debye characteristic temperature of 195°K[5] indicated

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[1] H. E. Nigh, S. Legvold, and F. H. Spedding, Phys. Rev. **132**, 1092 (1963).

[2] C. D. Graham, J. Appl. Phys. **36**, 1135 (1965).

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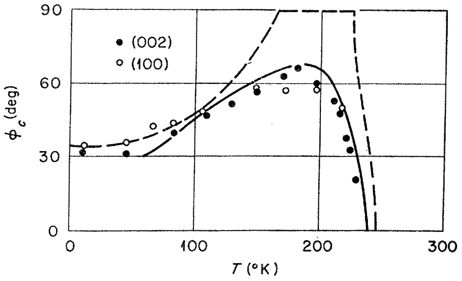

FIG. 3. Temperature dependence of ϕ_c, the angle between the moment orientation and the crystal c axis. The dotted curve represents torque measurements of Graham (Ref. 9) and the dashed curve those of Corner, Roe, and Taylor (Ref. 8).

a negligible thermal-motion correction for these reflections over this temperature range so that magnetic to nuclear intensity ratios could be obtained by direct comparison with the nuclear intensities above T_C. The thermal behavior of these magnetic to nuclear intensity ratios is shown in Fig. 1. For a ferromagnet this ratio can be expressed as:

$$\frac{I_{\text{mag}}}{I_{\text{nuc}}}=\frac{0.29q^2f(K)^2S^2(\sigma/\sigma_0)^2}{b_{\text{Gd}}^2}$$

in which q is the magnetic interaction vector, $f(K)$ is the magnetic form factor, $S=\frac{7}{2}$ for Gd^{+3}, σ/σ_0 is the relative magnetization and b_{Gd} is the nuclear scattering amplitude of Gd. If the angle between the moment direction and the crystal c axis is denoted by ϕ_c then

$$\langle q^2\rangle_{(100)}=1-\tfrac{1}{2}\sin^2\phi_c \qquad \text{(see Ref. 6)}$$

and

$$q^2{}_{(002)}=\sin^2\phi_c,$$

from which one notices that the (100) magnetic contribution is present regardless of the moment direction while the (002) intensity vanishes if $\phi_c=0$. The magnetic intensity of the (100) reflection vanishes at 294±2°K in good agreement with the T_C values obtained by extrapolation of the magnetization data.[1,2] The disappearance of the (002) magnetic intensity at 232°K indicates that $\phi_c=0$ from 232 to 294°K.

The d spacings of the (100) and (002) reflections are nearly equal and $f(K)^2\simeq 0.725$ [7] for both reflections. Therefore,

$$\frac{I_{\text{mag}}}{I_{\text{nuc}}}(100)+\frac{1}{2}\frac{I_{\text{mag}}}{I_{\text{nuc}}}(002)=\frac{2.58(\sigma/\sigma_0)^2}{b_{\text{Gd}}^2}$$

for any moment orientation. This sum is shown in Fig. 2 for coincident-temperature data. Included in the figure are squared relative spontaneous magnetization[2] and $B_{7/2}$ curves normalized to the 10°K data point. The adequate representation of the data by the $(\sigma/\sigma_0)^2$ curve indicates ferromagnetism throughout the entire temperature range. From the saturation value a Gd nuclear scattering amplitude of 0.95×10^{-12} cm is obtained. This assumes $S=\frac{7}{2}$, i.e., a localized moment of 7 μ_B rather than the 7.55 μ_B obtained from the saturation magnetization. This choice was made since the additional moment is usually attributed to conduction-electron polarization and as such would not show up in the Bragg reflections because of a rapidly decreasing form factor.

With b_{Gd} and $(\sigma/\sigma_0)(T)$ thus established, $\phi_c(T)$ can be obtained from the intensity variation of the individual reflections. This result is shown in Fig. 3 along with curves representing the torque measurements of Corner, Roe, and Taylor,[8] and Graham.[9] According to the present results the moment direction is parallel to the c axis from T_C to 232°K, moves away from that axis to a maximum deviation of about 65° near 180°K, and then back to within 32° of the c axis at low temperatures. From counting statistics only, ϕ_c values taken from the (002) data have an uncertainty of $\pm2\frac{1}{2}°$ while those from the (100) data are valid to $\pm4\frac{1}{2}°$. These ϕ_c values are in good agreement with the corrected data of Corner, Roe, and Taylor[8] and are somewhat smaller than Graham's values.

It is clear that all of the expected magnetic scattering appears at the normal lattice positions. Furthermore, there was no apparent broadening of the rocking curves below T_C whereas a spiral turn angle >2° would have produced a noticeable broadening. Also, there was no indication of a field dependence of the intensities in the range 0.1 to 5 Oe. We conclude that Gd is spontaneously ferromagnetic throughout the entire ordered region.

ACKNOWLEDGMENT

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