Some Magnetic Properties of Gadolinium Metal*

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The saturation magnetization of gadolinium has been found to follow a $T^{\frac{3}{2}}$ law between 20–250°K. The saturation magnetization at absolute zero, determined by extrapolation is 253.6±0.9 cgs units, corresponding to 7.12 Bohr magnetons. The data fall considerably below the theoretical curve predicted by the Debye quantum modification of the Weiss theory of ferromagnetism for the $^8S_{7/2}$ state. Curie point determinations have also been made.

I. INTRODUCTION

THE ferromagnetic properties of metallic gadolinium were reported in 1935 by Urbain, Weiss, and Trombe,[1] and by Trombe.[2] The value of the absolute saturation magnetization was cited as 253.5 cgs units, and the Curie point as 16°C. The development of processes for the separation of pure rare earth salts[3] and for the preparation of the pure metals[4] at this laboratory has made it possible to pursue such measurements further.

Preliminary measurements were made using a ballistic method with the sample in the form of a torus.[5] Since there appeared to be certain discrepancies between these preliminary measurements and those reported by Trombe, it was deemed desirable to extend these measurements to higher fields. This paper is a report on the extension of the preliminary measurements.

II. EXPERIMENTAL PROCEDURE

The spectrographic analysis of the gadolinium metal used was as follows: Ca, detectable, but less than 200 ppm; Sm, detectable, but less than 600 ppm; Fe, Co, and other rare earths, not detected.

Curie point determinations were made with annealed and unannealed samples. Magnetic moment measurements were made on an annealed sample only. The samples were annealed for twenty-four hours at 400°C.

The magnet used in these measurements was a 20-kw Weiss type electromagnet which was constructed at this laboratory. This magnet has a 12-inch core which tapers to six-inch pole faces. It is capable of producing a 20-kilogauss field over a $1\frac{1}{2}$-inch pole gap.

Magnetic moments were measured by a method similar to that suggested by Sucksmith.[6] The gradient of the electromagnet was calibrated using a sample of 99.99 percent pure iron, supplied by Johnson, Matthey and Company, Limited. The saturation magnetization at 20°C, for this sample, was taken to be 217.8 cgs units. The calibration was checked using a sample of nickel 99.97 percent pure and assuming a saturation value of 54.65 cgs units.

The calibration samples and the gadolinium were in the form of cylinders, 10 mm long and 1 mm in diameter. The magnetizing field was calculated using the expression

$$H=H_0-NI, \tag{1}$$

where H = the magnetizing field, H_0 = the applied field, I = the magnetization per cm^3, and N = the demagnetizing constant, taken to be 0.0875 for samples of the above aspect ratio.[7]

Two methods were used for the Curie point determination. In the preliminary measurements with the torus, the variation of magnetic moment with temperature was observed for a small constant field. The second method was to observe the variation of the spontaneous magnetization with temperature, using fields produced by the electromagnet, where the spontaneous magnetization was determined by the extrapolation technique of Weiss and Forrer.[8]

The cryogenic apparatus for the saturation magnetization measurements and the high field Curie point determinations consisted of a double Dewar. The inner Dewar contained the sample the temperature of which was controlled by a stream of helium gas. The outer Dewar contained a heat exchanger and a liquid bath. Helium gas was cooled in the exchanger, then mixed with a stream of warm helium before entering the inner Dewar. The temperature range from 39°–85°K was obtained with a hydrogen bath, from 85°–260°K with nitrogen, 200°–280°K with acetone and dry ice, 280°–310°K with ice water. The temperature was controlled to within a degree for saturation measurements and to within a quarter of a degree for Curie point determinations.

The temperature was measured by a copper-constantan thermocouple calibrated over the temperature range employed.

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[6] W. Sucksmith, Proc. Roy. Soc. (London) **A170**, 551 (1939).
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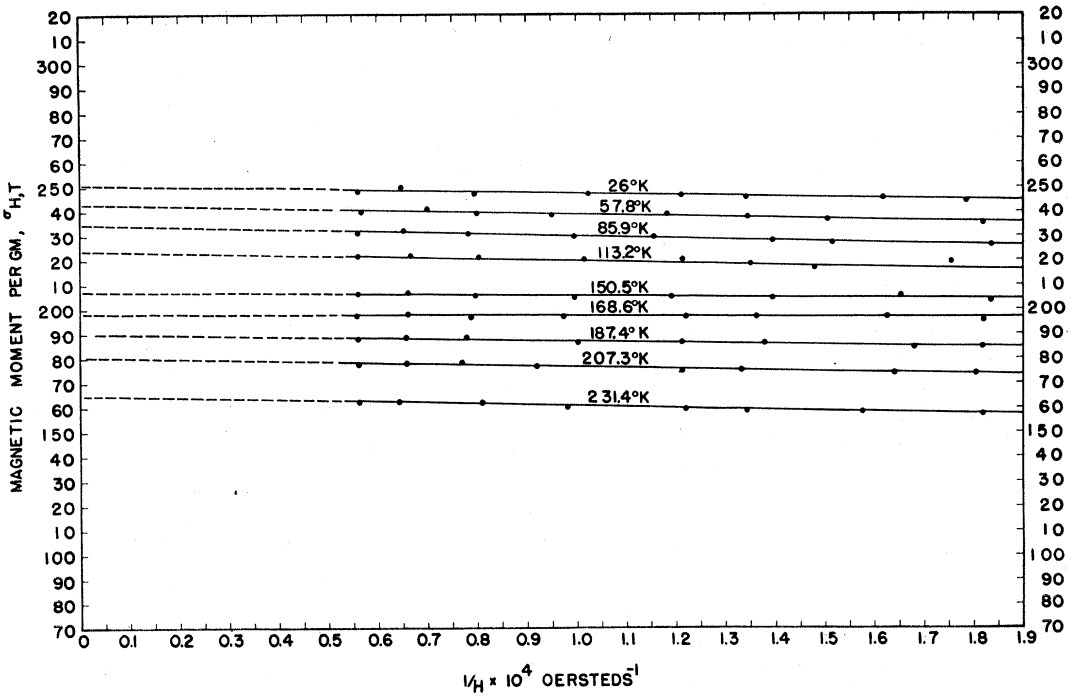

FIG. 1. Representative curves of the isothermal variation of magnetic moment of gadolinium as a function of $1/H$.

This apparatus was used throughout, except for the 20.4°K data. Here the sample was immersed directly in the hydrogen bath, and the necessary buoyancy corrections made.

III. RESULTS

Figure 1 shows several of the representative curves of the isothermal variation of the magnetic moment as a function of $1/H$. It was found that the variation, for magnetizing fields of 4000–18 000 oersteds over the temperature range of 20–253°K, is well represented by the expression

$$\sigma_{H,T}=\sigma_{\infty,T}(1-a/H), \tag{2}$$

where $\sigma_{H,T}$ = the magnetic moment per gram, $\sigma_{\infty,T}$ = the saturation moment per gram at T°K, H = the magnetizing field, and a = constant. The data were extrapolated to zero, using the method of least squares to find the saturation magnetization.

The values of the saturation magnetization, obtained by the above extrapolation, are plotted as a function of $T^{\frac{3}{2}}$ in Fig. 2(a). It may be observed that the data seem to agree quite well over the temperature range examined with an expression of the form,

$$\sigma_{\infty,T}=\sigma_{\infty,0}(1-bT^{\frac{3}{2}}), \tag{3}$$

where $\sigma_{\infty,0}$ = the absolute saturation magnetization, b = a constant, and T = absolute temperature. It may be further observed [Fig. 2(b)] that there appears to be some correlation between our data and the expression,

$$\sigma_{\infty,T}=\sigma_{\infty,0}(1-bT^{2}), \tag{4}$$

between 250°K and 130°K, but the data seem to fit the $T^{\frac{3}{2}}$ expression at least as well if not better than the T^2 expression over this range. Consequently, the $T^{\frac{3}{2}}$ law is used over the entire range for determining the absolute saturation magnetic moment.

Using again the method of least squares, the value of the saturation magnetization at absolute zero was found to be 253.6±0.6 cgs units. This saturation moment corresponds to 7.12 Bohr magnetons.

In Fig. 3 the ratio of $\sigma_{\infty,T}/\sigma_{\infty,0}$ is plotted as a function of the ratio of T/θ, where θ is the Curie temperature. Several curves, obtained from the Debye quantum modification of the Weiss theory of ferromagnetism for various J values, are also shown. To make com-

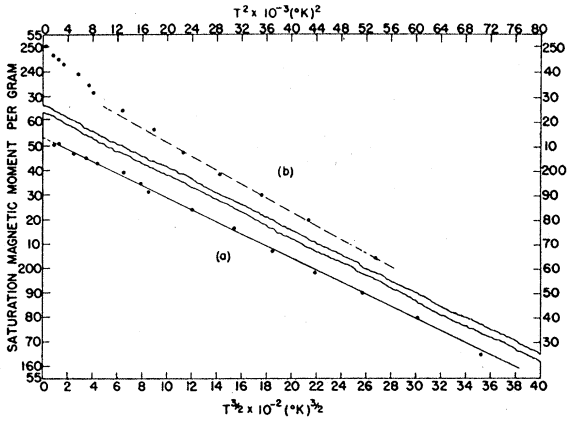

FIG. 2. (a) The saturation magnetic moment of gadolinium as a function of $T^{\frac{3}{2}}$; (b) of T^2.

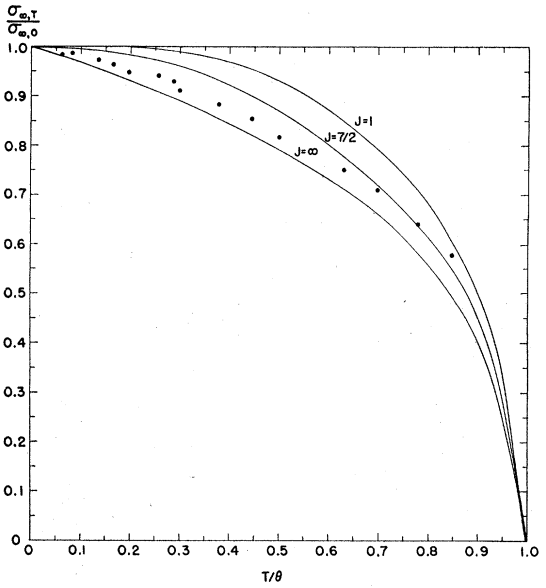

FIG. 3. The reduced magnetization curve for gadolinium; also, the theoretical curves for $J=1$, $J=7/2$ and $J=\infty$, from the Debye-Weiss quantum theory of ferromagnetism.

parison between experimental data and the Debye-Weiss theory, the ratio of $\sigma_{0,T}/\sigma_{0,0}$, where $\sigma_{0,T}$ is the spontaneous magnetization, should be plotted as a function of T/θ. However, our data indicate that the value of the spontaneous magnetization differs from the saturation magnetization by only a few percent, which would not change the location of the plotted points significantly.

The variation of the square of the spontaneous magnetization with temperature near the Curie point is shown in Fig. 4(b). The intersection of the extension of the nearly linear portion of this curve with the temperature axis was taken to be the Curie point, i.e., 17.7±0.3°C. In Fig. 4(a) the Curie point is determined from data obtained by the ballistic method using the torus and a small applied field (i.e., 23 oersteds) and was found to be 15.9±0.1°C.

IV. DISCUSSION OF RESULTS

Of particular interest is the fact that the value of the absolute saturation magnetization obtained by Trombe by extrapolation of a T^2 plot agrees exceedingly well with the value obtained here using an extrapolation of a $T^{\frac{3}{2}}$ plot. This discrepancy was obvious even in our preliminary measurements where, for instance, our value for the saturation magnetization at 77°K was some 4 percent lower than that obtained by Trombe.

Of interest also is the fact that the saturation magnetization of gadolinium follows a $T^{\frac{3}{2}}$ law over nearly its entire ferromagnetic range. Gadolinium thus seems to be peculiar in this respect as other ferromagnetics follow a $T^{\frac{3}{2}}$ law only at very low temperatures.

Our investigation also showed that gadolinium is not as magnetically "hard" as indicated by Trombe, although considerably "harder" than iron. The constant (a) in Eq. (2) is an indication of this "hardness" and has the value of about 170 from our data. This is to be compared with Trombe's value of 1250. This difference of "hardness" between our samples and Trombe's was also indicated by our preliminary measurements at low fields,[4] using the ballistic method.

The Curie point found by the ballistic method, Fig. 4(a), agrees quite well with Trombe's value, where he has defined the Curie point as the temperature at which ferromagnetism disappears in a small field. The Curie point is found to be some two degrees higher when it is defined as the temperature at which spontaneous magnetization disappears. The disagreement between the values of the Curie points does not seem unreasonable when the differences in their definitions are considered.

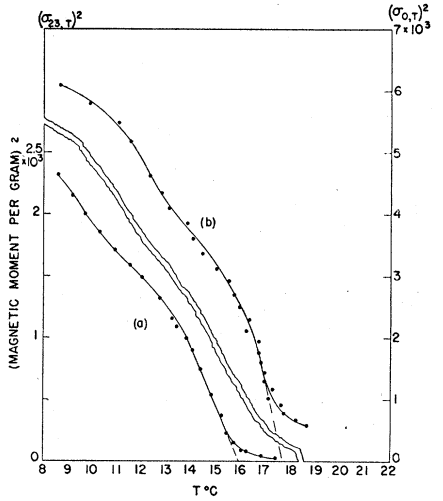

FIG. 4. (a) The Curie point of gadolinium using the ballistic method and an applied field of 23 oersteds. (b) The Curie point using spontaneous magnetization.

It was possible to repeat the data obtained by the ballistic method irrespective of whether the Curie point was approached from above or below, showing there was no temperature hystersis (within at least 0.1°) at the Curie point.

It may be observed that the absolute saturation magnetization is some 2 percent higher than would be predicted if only the spins of the $4f$ electrons contributed to the magnetization; this deviation is outside the experimental error. Also, the reduced magnetization data points fall far below the curve calculated for the ${}^8S_{7/2}$ spectrographic state using the Debye-Weiss theory of ferromagnetism. Had $\sigma_{0,T}$ data been used for this plot rather than $\sigma_{\infty,T}$ data, the experimental points would have been slightly lower than the points shown.

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