Magnetic Properties of Neodymium Single Crystals*

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Magnetic measurements have been made on single crystals of neodymium which were grown by the Bridgman technique. At 20.4°K and above, the susceptibility obtained with the field perpendicular to the c_0 axis of the hexagonal crystal is greater than that obtained with the field parallel to the c_0 axis. In this same temperature range no anisotropy is observed in the basal plane. At 4.2°K antiferromagnetic ordering is indicated by the data and magnetic anisotropy in the basal plane is observed.

I. INTRODUCTION

THE magnetic properties of polycrystalline neodymium have been investigated by Trombe,[1] by Klemm and Bommer,[2] by Elliott *et al.*,[3] and by Bates *et al.*[4] Trombe investigated the magnetic properties of neodymium over the temperature range 77°K to 297°K. These measurements showed a discontinuity in the slope of $1/\chi$ *vs* T in the neighborhood of 110°K. Above 110°K the effective magnetic moment was reported to be 3.59 Bohr magnetons, and below 110°K the effective moment was found to be 2.08 Bohr magnetons. Klemm and Bommer found the metal to obey a Curie-Weiss law from 90°K to 292°K, with an effective magnetic moment of 3.65 Bohr magnetons.

Elliott, Legvold, and Spedding have carried the measurements down to 20.4°K, using the Gouy method to measure the magnetic susceptibility. They also found a discontinuity in the slope of $1/\chi$ *vs* T at about 145°K. Above 145°K the effective magnetic moment was found to be 3.48 Bohr magnetons. Bates measured the susceptibility from 290°K to 1000°K and he found that it follows the Curie-Weiss law between 290°K and 500°K. He found an effective magnetic moment of 3.72 Bohr magnetons and -15°K as the paramagnetic Curie temperature.

II. SINGLE-CRYSTAL PREPARATION

Magnetic measurements were made on two single crystals of neodymium. These crystals were grown by the Bridgman method from the polycrystalline metal. The purification of the neodymium salts and their reduction are described elsewhere.[5,6] The crystals were grown under vacuum in a tantalum-wound resistance furnance. The crucible used was constructed of 5-mil tantalum sheet with arc-welded seams. The resulting material was found to consist of many single crystalline grains which could easily be seen by etching the sample with a solution of 1 to 2% nitric acid in absolute alcohol. Two of the large grains, about 3 mm on a cube edge, were removed with a jeweler's saw and etched to remove the cold-worked surface.

Neodymium has the hexagonal close-packed crystal structure with stacking of planes perpendicular to the c_0 axis of *abac*.[7,8] In this study the principal crystalline axes were located with the aid of x-rays and back-reflection Laue photographs. The crystals were mounted in a sample holder in such a way that the magnetic field could be directed along any desired crystalline axis.

The body force method of measuring the magnetic moment of the sample was used. The electromagnet is described in an earlier paper.[9] Various temperatures of the sample were obtained by boiling either liquid hydrogen or nitrogen and passing the gas over the sample. By controlling the amount of heat introduced into the gas stream before it reached the sample, temperatures from 25°K to room temperature could be obtained. Temperatures of 20.4°K and 4.2°K were obtained by immersing the sample in either liquid hydrogen or liquid helium.

Since both crystals were obtained from neighboring regions of the bulk material, only one spectroscopic analysis was obtained. The sample for the analysis was

TABLE I. The magnetic susceptibility per gram of neodymium for the field parallel and perpendicular to the c_0 axis.

T (°K)	$10^6\times\chi_g(\parallel)$	$10^6\times\chi_g(\perp)$
34.3	...	370.9
39.4	258.1	...
40.0	249.1	303.5
43.95	228.7	272.2
69.95	144.9	160.2
70.0	144.6	159.3
100.0	104.0	111.8
100.0	103.2	109.6
140.0	76.87	80.26
149.95	71.39	74.29
180.0	59.46	61.12
220.0	49.14	51.50
230.0	47.88	49.02
240.0	45.39	47.03
280.0	40.46	40.98

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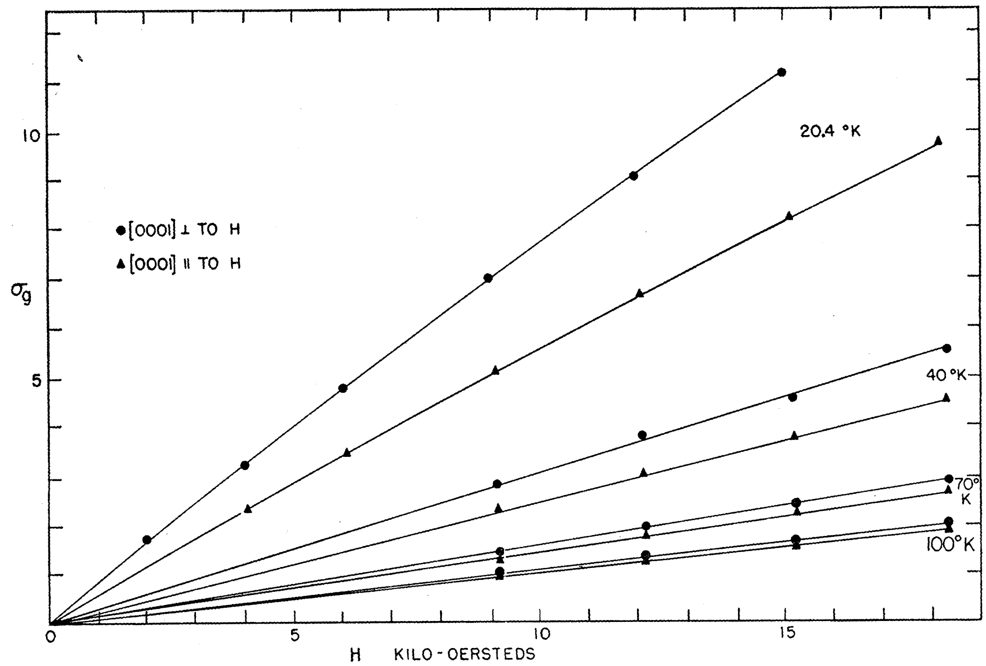

FIG. 1. Magnetic moment per gram of Nd *vs* field at low temperatures.

taken from the material adjacent to the crystals. The analysis showed the following impurities: Si, 0.1%; Ca, detectable but less than 0.03%; Fe and Mg detectable but less than 0.01%; Pr, Sm, Ta, and Cr not detected.

III. RESULTS

Figure 1 shows representative σ_g *vs* H isotherms obtained with the magnetic field parallel and perpendicular to the c_0 axis, where σ_g is the magnetic moment per gram. The data indicate the presence of magnetic anisotropy since $\chi_\perp$ is larger than $\chi_{\parallel}$ where $\chi_\perp$ and $\chi_{\parallel}$ are the susceptibilities with H perpendicular and parallel to c_0 respectively. The atomic magnetic moments apparently favor an alignment perpendicular to the c_0 axis. A careful attempt was made to look for the effects of anisotropy in the basal plane at both 20.4°K and 160°K. It was observed that within the 0.2% limit of

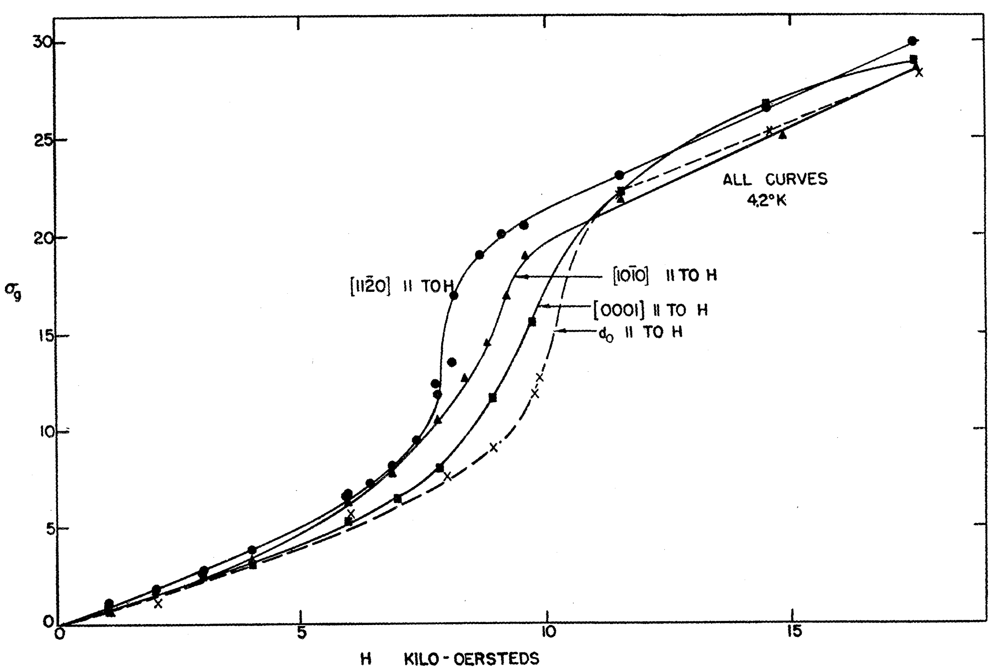

FIG. 2. Magnetic moment per gram of Nd *vs* field at 4.2°K.

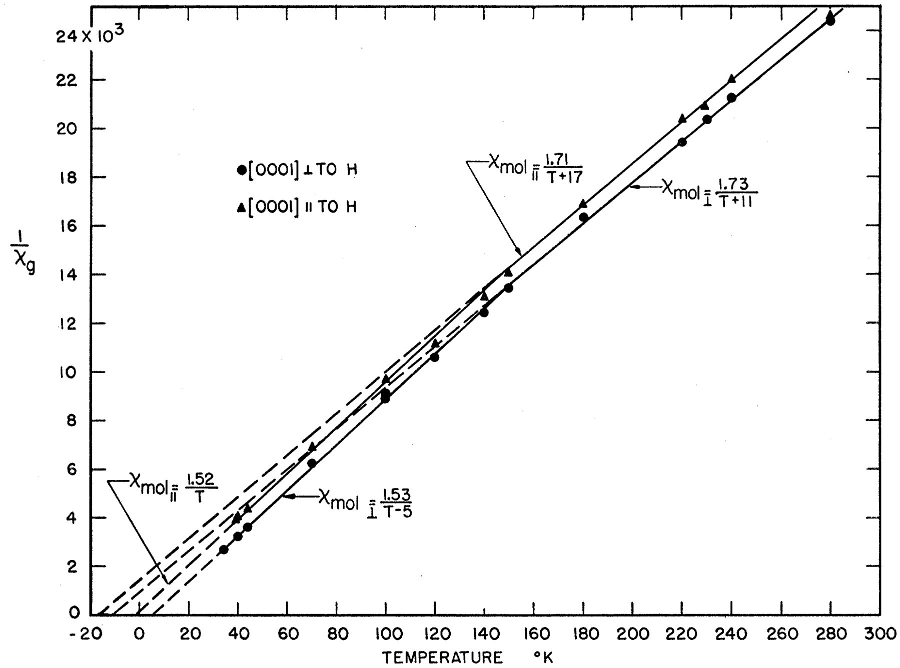

FIG. 3. Reciprocal of the susceptibility per gram of Nd *vs* temperature.

detectability the magnetic moment for a given field is isotropic in the basal plane.

Figure 2 shows the isotherms obtained at 4.2°K with the applied field along various crystalline directions. To describe the directions along which H is applied, unit cell vectors along the three a_0 axes and along the c_0 axis are used. Indices [$hkil$] designate the components of a direction vector along these unit vectors. Equivalent directions in the unit cell are obtained by permutations on h, k, i or $-h$, $-k$, $-i$. The direction labeled d_0 is contained in a plane which is defined by the [0001] or c_0 direction and any one of the [11$\bar{2}$0] directions, and d_0 makes an angle with the [0001] direction of 40±5°. For all the directions which were investigated, the crystal was in rotational equilibrium, i.e., the torques exerted on the crystal by the magnetic field were zero.

For all isotherms, except the ones at 20.4°K and 4.2°K, the magnetic moment was a linear function of the magnetic field. The susceptibilities per gram, χ_g, for temperatures above 20.4°K are given in Table I. Figure 3 is a plot of the inverse of the susceptibility per gram *vs* the absolute temperature. The data for either H parallel or H perpendicular to the c_0 axis fall on two straight lines intersecting at about 145°K.

In Table II is shown a comparison of the results of this study with those of other investigators. In order to compare the results when using single crystals with the polycrystalline results, the following expressions should be used: $C=C_{\parallel}=C_{\perp}$, and $\Delta=\frac{1}{3}\Delta_{\parallel}+\frac{2}{3}\Delta_{\perp}$. Here $C_{\parallel}$ and $C_{\perp}$ are, respectively, the Curie constants per mole obtained with the field parallel and perpendicular to the c_0 axis, and the quantities $\Delta_{\parallel}$ and $\Delta_{\perp}$ are the paramagnetic Curie points obtained with the field parallel and perpendicular to the c_0 axis. The theoretical μ_{eff} for the $^4I_{9/2}$ state of the tripositive Nd ion is 3.68.

TABLE II. Comparison of the results of this study with the data of other investigators.

Investigator	Curie constant per mole (°K/mole)	μ_{eff} (Bohr magnetons)	Curie point (°K)	Temperature range (°K)
Klemm and Bommer[a]	1.664	3.65		90 –292
Trombe[b]	1.609	3.59		110 –297
	0.540	2.08		77 –110
Elliott, Legvold,	1.695	3.68	−16	145 –300
and Spedding[c]	1.507	3.48	1	31.5–145
Bates[d]	1.724	3.72	−15	290 –500
This study	1.720±0.03	3.71±0.05	−15±2	145 –280
	1.52 ±0.02	3.45±0.04	3±2	35 –145

[a] See reference 1. [b] See reference 2. [c] See reference 3. [d] See reference 4.

It appears from the data that neodymium is antiferromagnetic at 4.2°K or that it is at least ordered magnetically. This view is supported to some extent by the data of Parkinson, Simon, and Spedding,[10] who have measured the heat capacity of neodymium from 2°K to 160°K. They observed two rather sharp peaks in the specific heat, one near 19°K and the other near 7.5°K.

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