

Electrical Resistivity of the Heavy Rare-Earth Metals*

R. V. Colvin[†], Sam Legvoid and F. H. Spedding

Institute for Atomic Research and Department of Physics

Iowa State University, Ames, Iowa

Abstract--The electrical resistivities of polycrystalline Gd, Tb, Dy, Ho, Er, Tm, and Lu have been measured between 1.3°K and room temperature. The effects of magnetic ordering processes are apparent in the resistivities of all these elements except Lu.

I. INTRODUCTION

We present here the temperature dependence of the electrical resistivities of Gd, Tb, Dy, Ho, Er, Tm and Lu. All of these elements have the hexagonal close-packed crystal structure¹ and have very similar physical and chemical properties. For a given member of this group good correlations are found in the abnormal temperature dependence of such physical properties as specific heat, magnetic susceptibility, thermal expansion, thermoelectric effect, and electrical resistivity.

The measurements reported below were made on the best samples presently available in the Ames Laboratory. For Gd, Dy, and Er we believe these samples are superior to samples for which resistivities

* Contribution No. 893. Work was performed in the Ames Laboratory of the U. S. Atomic Energy Commission.

[†] Now at the Edgar C. Bain Laboratory of the U. S. Steel Corporation.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

were reported previously.² This is manifested in more distinct breaks in the resistivity curves, lower values of the resistivity and smaller slopes in the curves. In other earlier work Bridgman³ reported the resistivities of a number of rare-earth metals from 0°C to room temperature. The resistivities of single crystals of Dy⁴ and Er⁵ have been measured and polycrystalline results have been predicted from these measurements. Agreement with the results reported here is good.

II. EXPERIMENTAL PROCEDURE

A. Apparatus

The variable temperature apparatus used for this investigation is shown in Fig. 1. A similar heat leak chamber was used and described by Andersen.⁶ Appropriate changes in the apparatus were made to permit operation with liquid hydrogen and nitrogen as well as with liquid helium. Since operation of the apparatus at the higher temperatures was not economically feasible when helium was used as the cold reservoir, a nitrogen reservoir, which was refilled automatically, was used to maintain any temperature between 77.5°K and 400°K for indefinite periods of time.

Temperatures were changed by using pulses of electrical power in the 140 ohm heater coil to obtain the temperature desired. Automatic temperature control was used to maintain a temperature when it was desirable to do so. It was found that in the temperature regions where the resistivity was well behaved it was sufficient to have

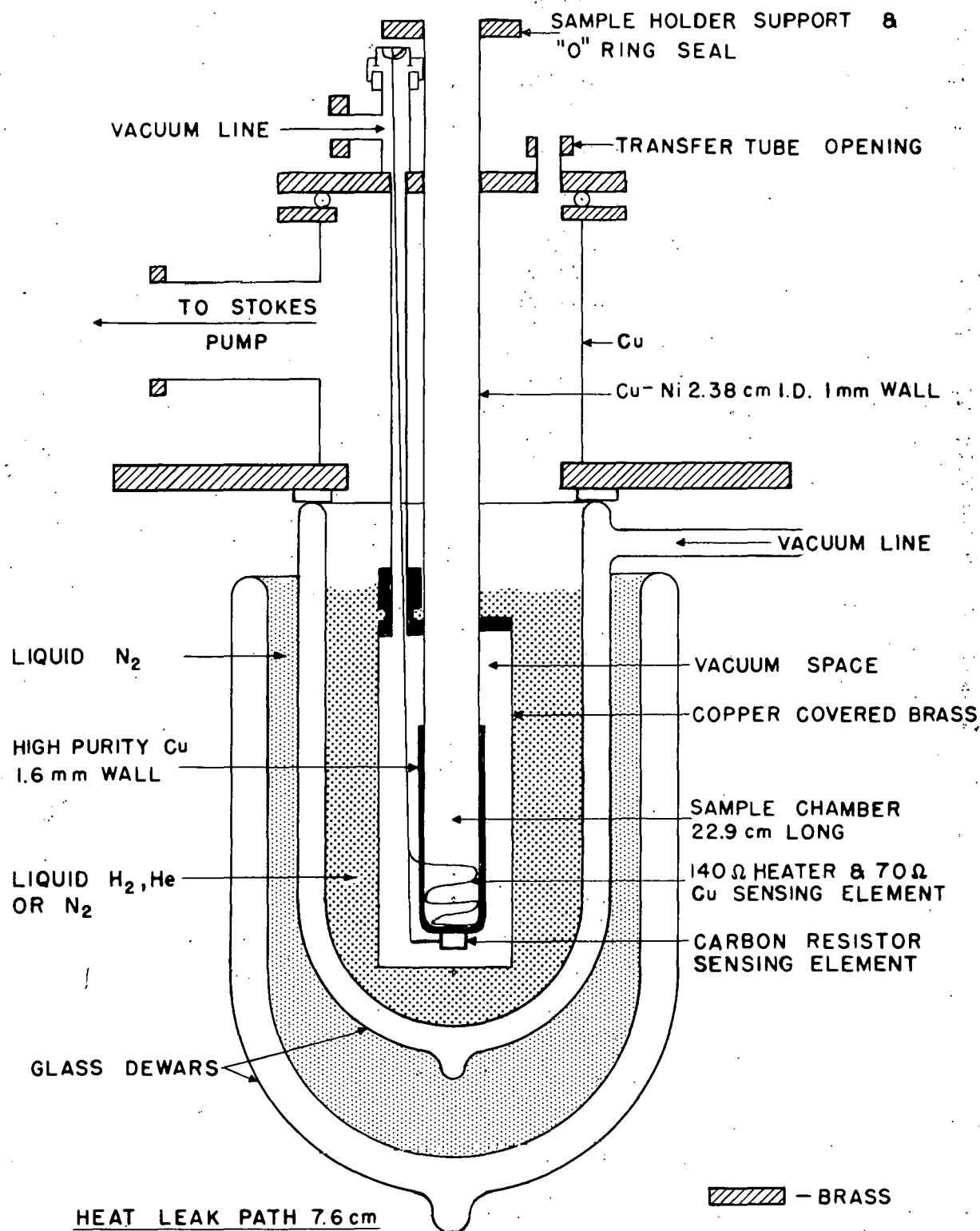


Fig. 1. Low temperature heat leak chamber.

approximate equilibrium conditions to obtain good data. Temperatures and resistivities were measured simultaneously.

The sample resistance was determined by the usual four-probe method using current reversal. An electronic constant-current power supply was used to supply .342 amp to all of the samples measured. Sharpened brass wedges approximately an inch apart mounted on a quartz block were used as potential contacts. Sample diameters were determined with a micrometer.

Most of the measurements below 20°K were made in the liquid hydrogen or liquid helium baths. Temperature measurements were made with a vapor pressure thermometer or with copper constantan thermocouples.

The probable error in the determination of the sample resistivities was about 1%. Temperatures were known to the nearest .1°K.

B. Samples

All of the samples were cast rods 3/16-in. in diameter and 2-in. long. The curves for Gd, Tb, Tm and Lu were obtained after annealing. Annealing attempts on Ho and Er were unsuccessful. The curves reported for these two elements were obtained prior to annealing. No attempt was made to anneal Dy.

It has been observed that Ta impurities in a rare-earth metal will greatly reduce the resistivity and give erroneous results below the superconducting transition of Ta. Consequently, we have not plotted any points below 4.4°K for Tm and Lu which apparently pick up Ta in the casting process. For the same reason we have taken the residual resistivity to be the 4.4°K value for all the samples. On the basis of the measurements made on many samples and their extensive analyses we can find no contradiction to the commonly accepted proposal that the residual resistivity of metals is related to the over-all sample purity.

In Table I we show the results of spectrographic and other tests for impurities in the samples used in this work.

III. RESULTS

Antiferromagnetism is apparently characterized by a peak in the resistivity curve. The transition from ferromagnetism to antiferromagnetism is identified with a jump in resistivity. A well-behaved resistivity curve for Lu was anticipated since it has a filled 4f shell.

The electrical resistivities observed on the heavy rare-earth metals are shown in Figs. 2-8 and are discussed below:

Gadolinium—A sharp change in slope occurs in the resistivity of Gd between 291 and 292°K (Fig. 2). Measurements of magnetic

Table I. Sample analysis

Element	Analysis
Gadolinium (not distilled)	^d Dy $\bar{<}$.01; Y $\bar{<}$.05; Pb $\bar{<}$.01; Eu $\bar{<}$.01; Sm $\bar{<}$.05; Nd $\bar{<}$.05; Mg $\bar{<}$.01; Si, .025; Ca, .06; Fe, .02; Ta $\bar{<}$.1; C, 210 ppm; N, 130 ppm; Ag, Al, As, Au, B, Be, Bi, Co, Cr, Cu, Ga, Ge, Hg, Ir, Mn, Mo, Na, Pb, P, Pd, Rh, Ru, Sn, Sr, Ti, Tl, V, W, Zn, Zr not detected; Ni-trace.
Terbium (not distilled)	Dy, 0.05; Gd, 0.05; Ca, 0.04; Eu, Fe, Ho, Si, Ta, Y, Yb, Tm, La, Lu, Nd, Pr, Er, not detected.
Dysprosium (distilled)	Y $\bar{<}$.01; Tb $\bar{<}$.1; Yb $\bar{<}$.005; Er $\bar{<}$.02; Ho $\bar{<}$.02; Ta, 0.2; Fe $\bar{<}$.01; Si $\bar{<}$.03; Ca $\bar{<}$.05; C, 100 ppm; N, 15 ppm; Mg very faint trace.
Holmium (distilled)	^d ^d Tm $\bar{<}$.01; Er $\bar{<}$.02; Dy $\bar{<}$.04; Y $\bar{<}$.01; Ta, .2; Fe, .01; Ca, .05; Si $\bar{<}$.02; C, 75 ppm; N, 94 ppm; Ni, trace; Cu, trace; Mo, Cr, Al, Sc not detected.
Erbium (distilled)	Ca, $\bar{<}$.01; Fe, .02; Mg $\bar{<}$.01; Si $\bar{<}$.01; Y $\bar{<}$.01; Dy $\bar{<}$.005; Yb $\bar{<}$.0002; Tm $\bar{<}$.002; Ho $\bar{<}$.008; Ca, faint trace; Cu, trace; La, trace.
Thulium (distilled)	^d Lu $\bar{<}$.003; Yb $\bar{<}$.0005; Er $\bar{<}$.004; Ho $\bar{<}$.04; Y $\bar{<}$.02; Ca $\bar{<}$.05; Mg $\bar{<}$.05; Fe, .02; Si $\bar{<}$.01; C, 120 ppm; N, 9 ppm; Ta, 1.5; Ag, As, Au, Ba, Bi, Co, Ge, Hf, Hg, In, Mo, Na, Nb, Ni, Te, Ti, Tl, V, W, Zn, Zr, not detected. Cu, trace; Al trace; Mn, trace; Cr, trace.
Lutetium (not distilled)	^d ^d Y $\bar{<}$.05; Sc $\bar{<}$.02; Yb $\bar{<}$.005; Tm $\bar{<}$.002; Ca $\bar{<}$.05; Mg $\bar{<}$.03; Cr $\bar{<}$.02; Fe $\bar{<}$.01; Si $\bar{<}$.03; C, 66 ppm; N, 720 ppm; As, Au, Ba, Be, Cd, Co, Cr, Hf, Hg, In, Na, P, Pt, Ru, Te, Tl, V, Zr not detected, Cu, trace; Mn, trace; Nb, trace; Ni, trace; Sn, trace; Ta (strong line).

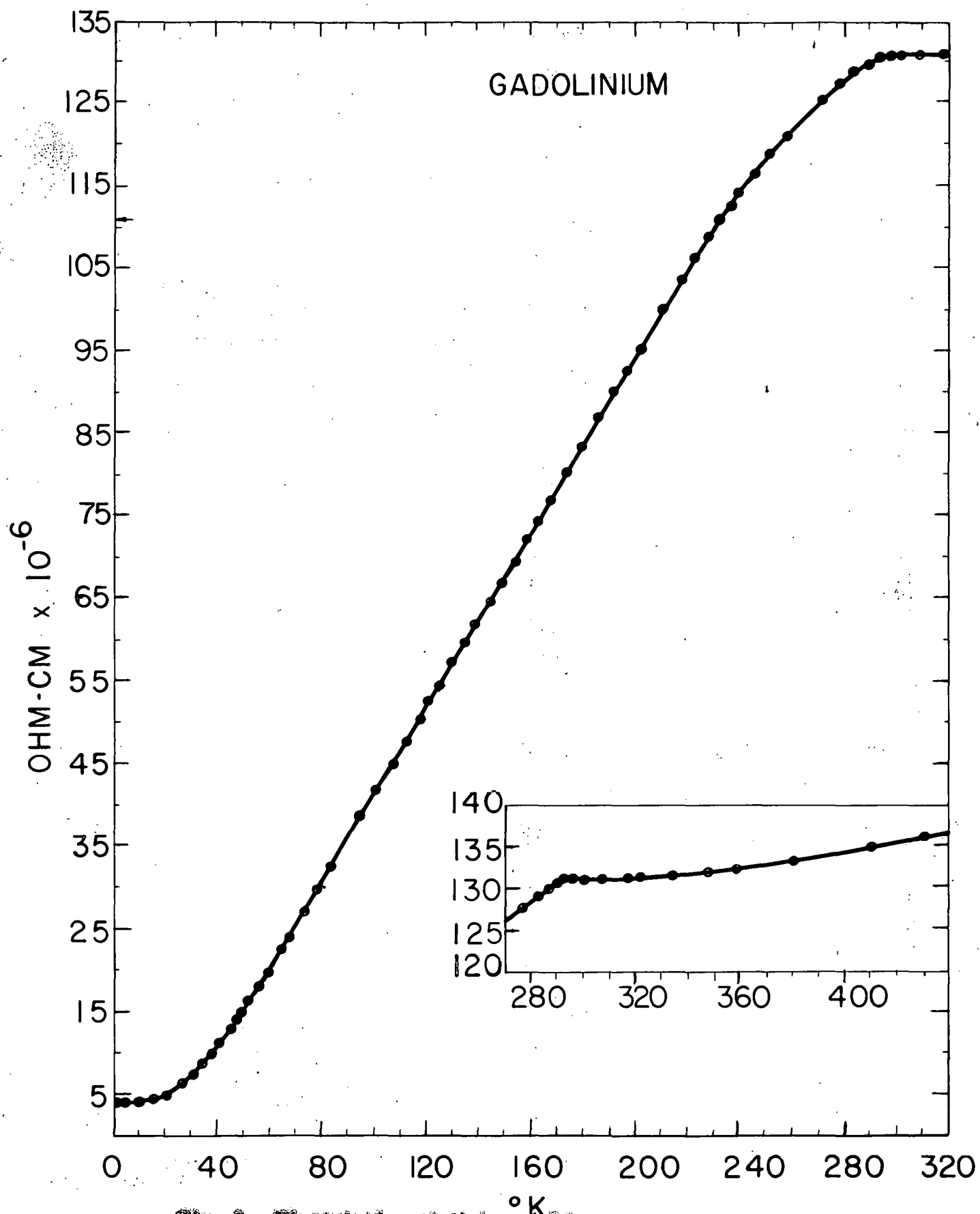


Fig. 2. Electrical resistivity of Gd vs temperature.

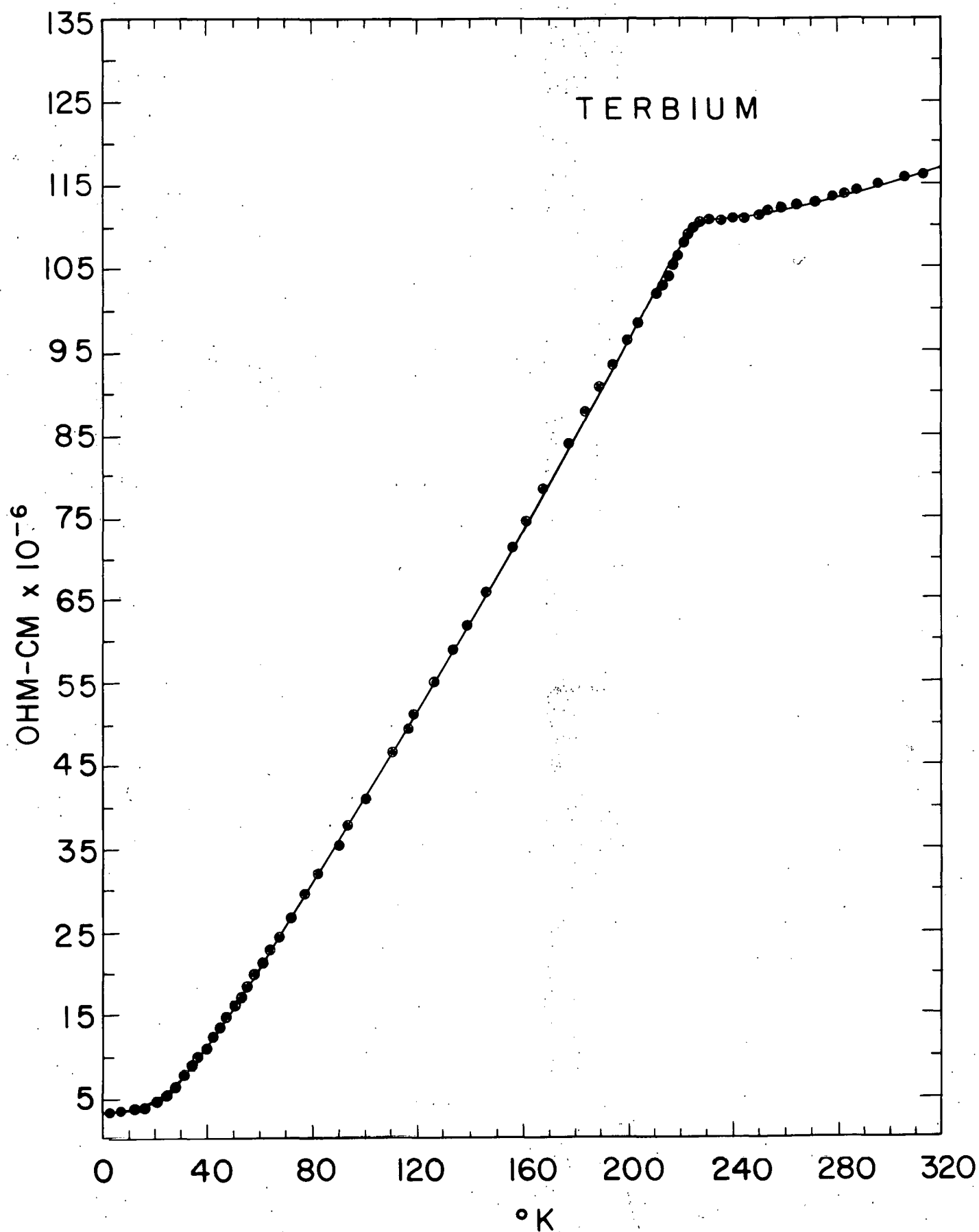


Fig. 2. Electrical resistivity of Tb vs temperature.

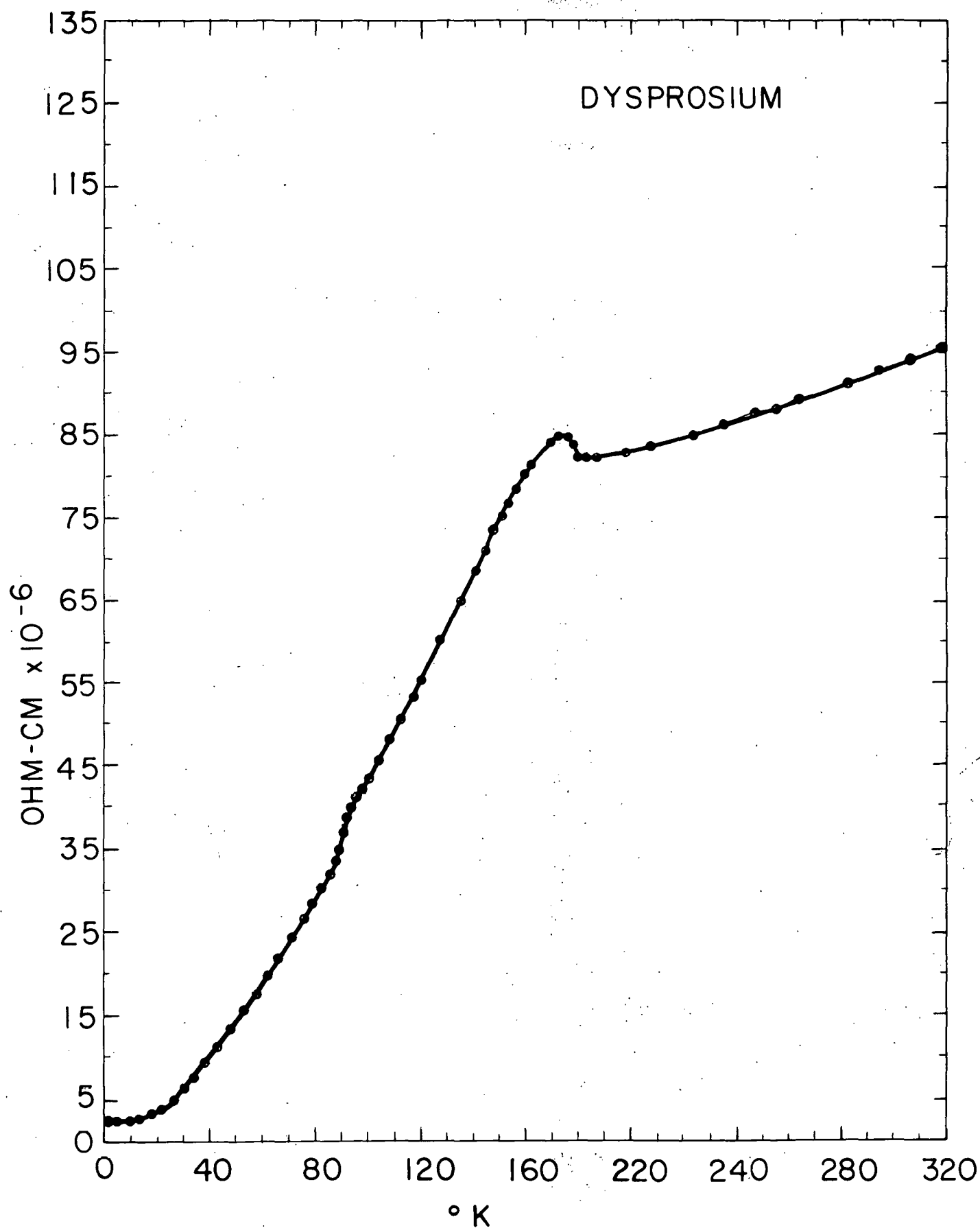


Fig. 6. Electrical resistivity of Dy vs temperature.

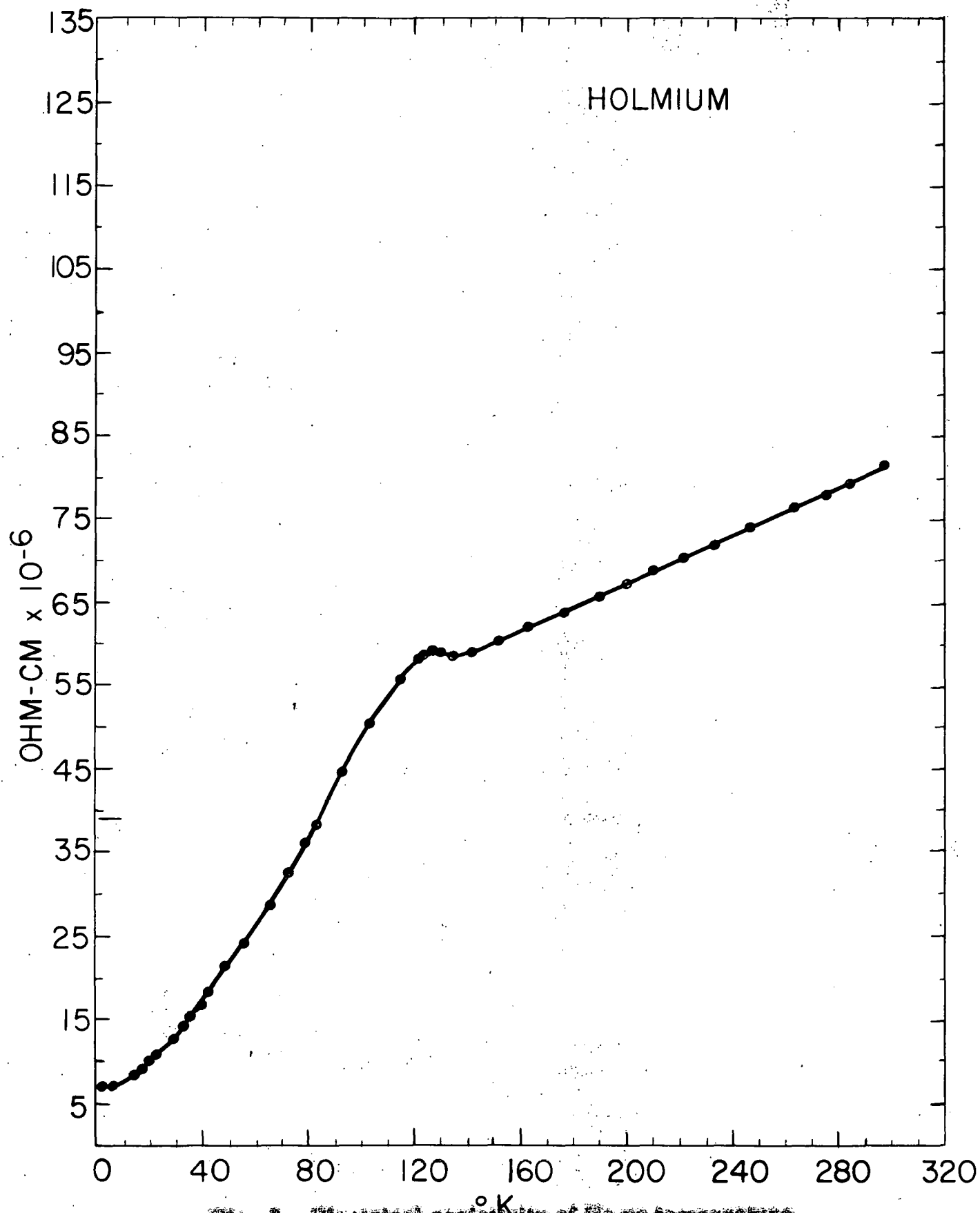


Fig. 5. Electrical resistivity of Ho vs temperature.

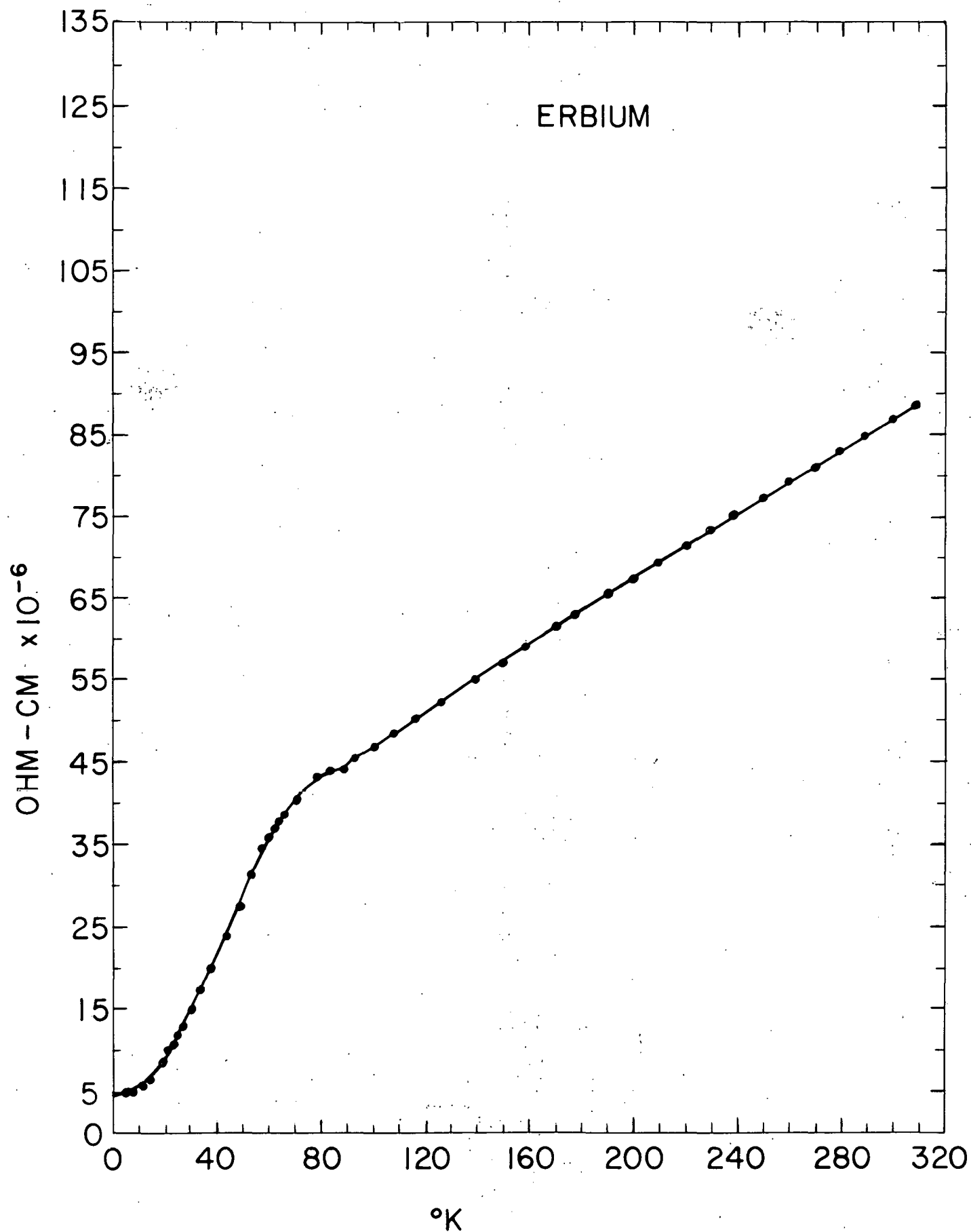


Fig. 6. Electrical resistivity of Er vs temperature.

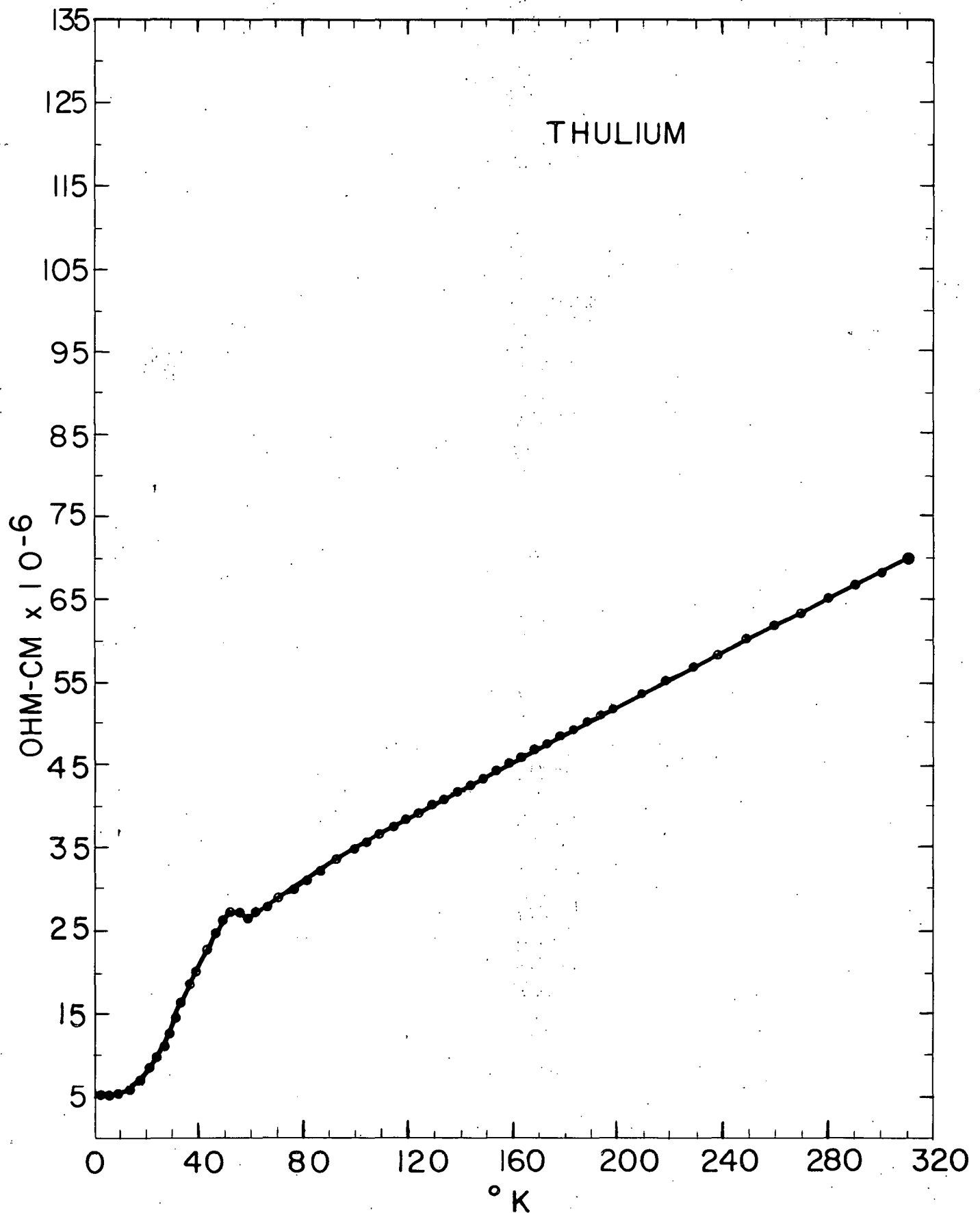


Fig. 7. Electrical resistivity of Th vs temperature.

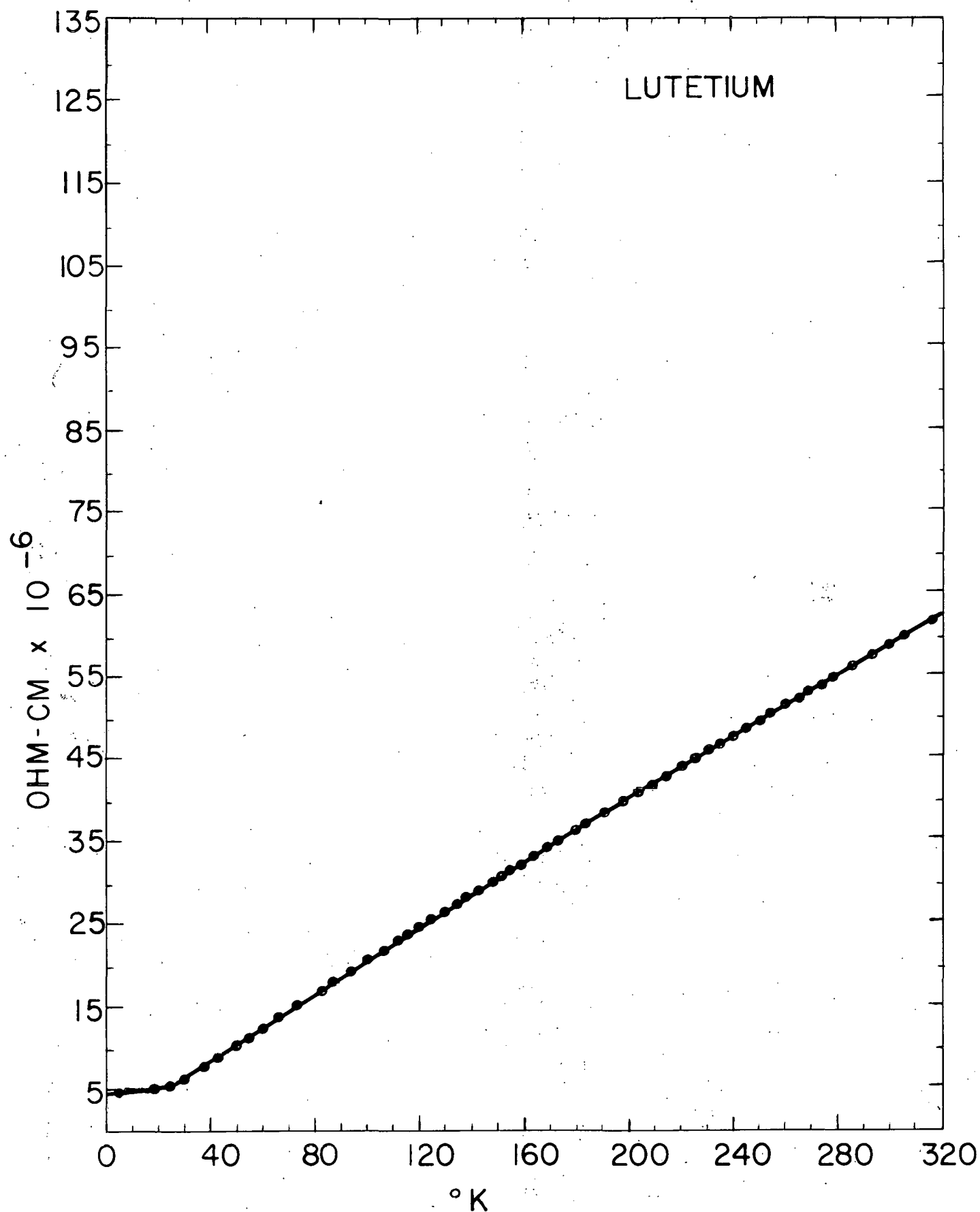


Fig. 8. Electrical resistivity of Lu vs temperature

moments,² thermal expansion,⁷ thermoelectric effect⁸ and specific heat⁹ indicate abnormal behavior near this temperature and suggest that the sharp change in slope should be associated with ferromagnetic ordering.

Terbium—There is a sharp change in the slope of the resistivity curve for terbium at 229°K as seen in Fig. 3. A careful study of the resistivity just below this temperature showed a slight increase in slope with increasing temperature at 219°K. Measurements of magnetic moments,¹⁰ specific heat,¹¹ thermal expansion⁷ and thermoelectric effect⁸ suggest that the region from 219°K to 229°K is an antiferromagnetic state and the region below 219°K is ferromagnetic.

Dysprosium—As seen in Fig. 4, a sharp increase in resistivity with increasing temperature occurs at 90°K. A peak was found at 174°K. An applied magnetic field of 22.4 kilo-oersteds will remove most of the peak and will suppress the resistivity jump at 90°K. Measurements of specific heat,¹² magnetic moments,^{13, 14} thermoelectric effect⁸ and thermal expansion⁷ suggest antiferromagnetism from 90°K to 174°K and ferromagnetism below 90°K.

Holmium—A change in slope (see Fig. 5) occurs near 19°K similar to the change at 90°K in Dy. A peak was found at 127°K. Measurements of specific heat,¹⁵ magnetic moments¹⁶ and thermoelectric effect⁸ suggest ferromagnetism below 19°K and antiferromagnetism from 19°K to 127°K.

Erbium—A pronounced minimum occurs in the curve for erbium at 80°K as seen in Fig. 6. All the points were taken with the temperature increasing from 4.2°K. Measurements of magnetic moments^{2, 5, 17} and specific heat¹⁸ suggest that the metal is ferromagnetic below 20°K and antiferromagnetic between 20° and 80°K. The ferro-antiferro transition is not detectable in the polycrystalline resistivity curve reported here.

Thulium—A peak occurs at 54.5°K. Measurements of specific heat¹⁹ and magnetic moments¹⁶ suggest T_m is antiferromagnetic below 54.5°K. The resistivity data below this temperature do not indicate a change to a ferromagnetic state.

Lutetium—The resistivity of Lu was found to be well behaved over the temperature region investigated. Measurements of specific heat¹⁹ and thermoelectric effect⁸ also indicate normal behavior for a metal.

IV. MAGNETIC DISORDER RESISTIVITY

The proposal by Kasuya²⁰ that the electrical resistivity of rare-earth metals is not due entirely to impurity and phonon scattering of conduction electrons appears to be a reasonable suggestion consistent with the experimental results. The manner in which one might calculate the magnetic contribution to the resistivity is not clearly established.

We have treated the best data available according to the proposals of Anderson and Legvold²¹ for calculation of the magnetic disorder

contribution to the resistivity. All of the results used for these calculations with the exception of Tb were obtained after the publication of the letter by Anderson and Legvold.²¹ In Table II we have tabulated the residual resistivities and our estimates of the magnetic contribution to the resistivity of each element. In Fig. 9 we have plotted these contributions as functions of two different parameters. The $S(S+1)$ parameter is the one used by Anderson and Legvold²¹ and the parameter $\frac{J+1}{J} S^2$ has been suggested by Brout and Suhl.²²

Table II. Magnetic and Residual Resistivities

Element	ρ_{mag}	ρ_{res}
Gd	106.9	4.4
Tb	86.4	3.5
Dy	58.5	2.4
Ho	33.5	7.0
Er	25.2	2.4
Tm	16.2	5.6
Lu	0.2	4.5

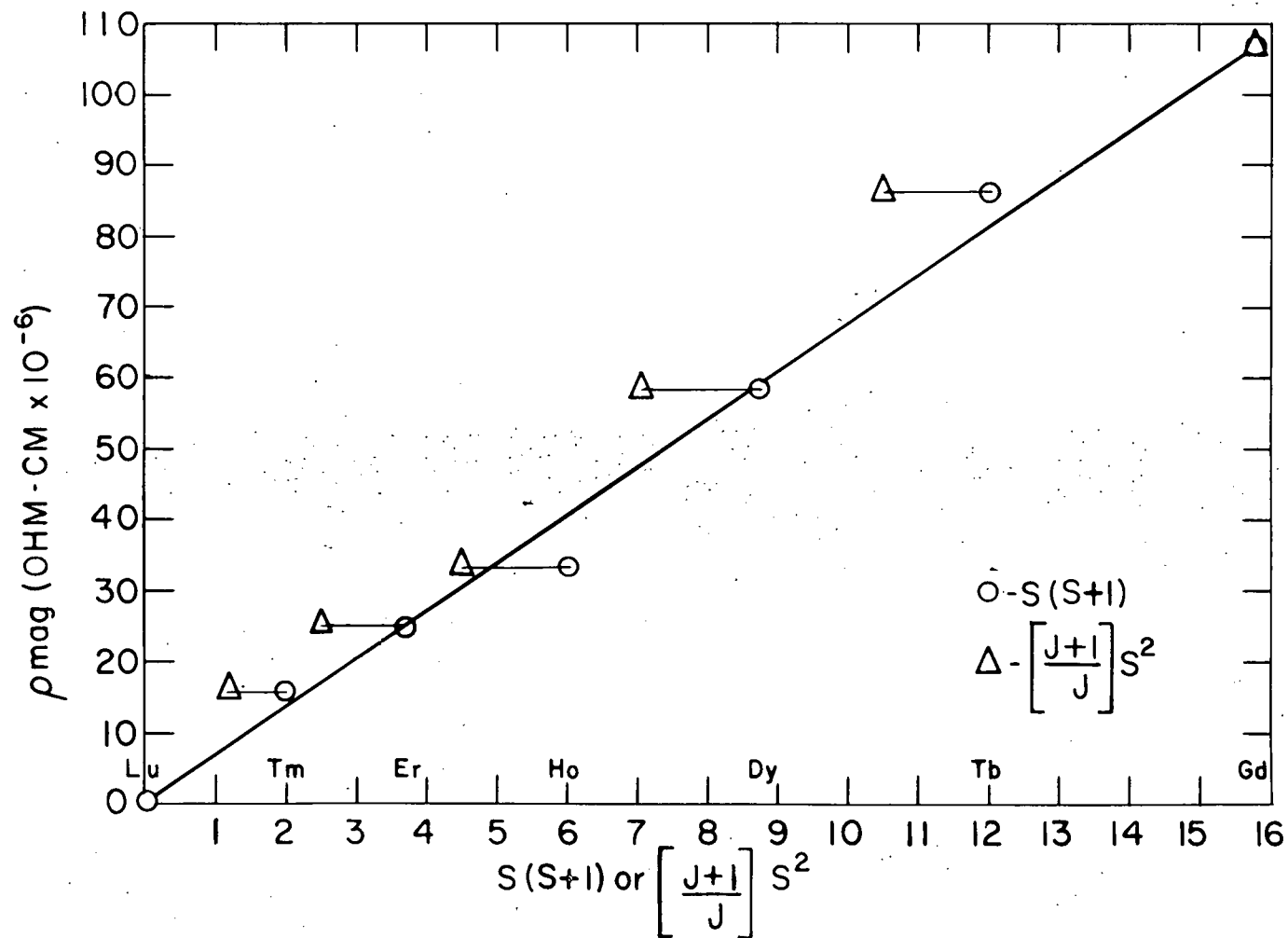


Fig. 7. The magnetic disorder contribution to the electrical resistivity vs $S(S+1)$ and $\left[\frac{J+1}{2} \right] S^2$ for the heavy rare earths, Gd to Lu.

V. ACKNOWLEDGMENTS

The authors are indebted to Dr. J. Powell for preparing the pure rare-earth salts needed, and to Mr. G. Wakefield and Mr. C.

Habermann for producing the metal samples used. Mr. J. Alstad gave much valuable assistance in the treatment of data.

- (1) F. H. Spedding, S. Legvold, A. H. Daane and L. D. Jennings, Progress in Low Temperature Physics (North-Holland Publishing Company, Amsterdam, Netherlands, 1957), Vol. II.
- (2) S. Legvold, F. H. Spedding, F. Barson and J. F. Elliott, *Rev. Mod. Phys.* 25, 129 (1953).
- (3) P. W. Bridgman, *Proc. Am. Acad. Arts Sci.* 83, 1 (1954).
- (4) P. M. Hall, S. Legvold, and F. H. Spedding, *Phys. Rev.* 117, 971 (1960).
- (5) R. W. Green, Unpublished PhD thesis, Iowa State University Library, Ames, Iowa (1960).
- (6) G. S. Anderson, S. Legvold, and F. H. Spedding, *Phys. Rev.* 109, 243 (1958).
- (7) F. Barson, S. Legvold and F. H. Spedding, *Phys. Rev.* 105, 418 (1957).
- (8) H. Born, Ames, Iowa. Private communication (1959).
- (9) M. Griffel, R. E. Skochdopole and F. H. Spedding, *Phys. Rev.* 93, 657 (1954).
- (10) W. C. Thoburn, S. Legvold, and F. H. Spedding, *Phys. Rev.* 112, 56 (1958).
- (11) L. D. Jennings, R. M. Stanton and F. H. Spedding, *J. Chem. Phys.* 27, 909, (1957).
- (12) M. Griffel, R. E. Skochdopole and F. H. Spedding, *J. Chem. Phys.* 25, 75 (1956).
- (13) J. F. Elliott, S. Legvold and F. H. Spedding, *Phys. Rev.* 94, 1143 (1954).
- (14) D. R. Behrendt, S. Legvold and F. H. Spedding, *Phys. Rev.* 109, 1544 (1958).

- (15) B. C. Gerstein, M. Griffel, L. D. Jennings, R. E. Miller, R. E. Skochdopole and F. H. Spedding, J. Chem. Phys. 27, 394 (1957).
- (16) B. L. Rhodes, S. Legvold and F. H. Spedding, Phys. Rev. 109, 1547 (1958).
- (17) J. F. Elliott, S. Legvold and F. H. Spedding, Phys. Rev. 100, 1595 (1955).
- (18) R. E. Skochdopole, M. Griffel and F. H. Spedding, J. Chem. Phys. 23, 2258 (1955).
- (19) L. D. Jennings, Private communication. (To be published in J. Chem. Phys.)
- (20) Taduo Kasuya, Prog. Theoret. Phys., Kyoto, Japan 16, 58 (1956).
- (21) G. S. Anderson and Sam Legvold, Phys. Rev. Letters 1, 322 (1958).
- (22) R. Bront and H. Suhl, Phys. Rev. Letters 2, 387 (1959).

Figure Captions

- Fig. 1. Low temperature heat leak chamber.
- Fig. 2. Electrical resistivity of Gd vs temperature.
- Fig. 3. Electrical resistivity of Tb vs temperature.
- Fig. 4. Electrical resistivity of Dy vs temperature.
- Fig. 5. Electrical resistivity of Ho vs temperature.
- Fig. 6. Electrical resistivity of Er vs temperature.
- Fig. 7. Electrical resistivity of Tm vs temperature.
- Fig. 8. Electrical resistivity of Lu vs temperature.
- Fig. 9. The magnetic disorder contribution to the electrical resistivity vs $S(S+1)$ and $\frac{(J+1)S^2}{J}$ for the heavy rare earths, Gd to Lu.