

## Specific Heat of Lutetium between 3 and 25°K\*

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The specific heat of lutetium metal has been measured between 3 and 25°K. By assuming 11.27T mJ/mole °K for the electronic contribution, the Debye temperature  $\Theta_D$  has been calculated and is presented as a function of temperature.  $\Theta_D$  drops off quite rapidly from its 0°K value of 210°K, as is common in hcp metals, and reaches a minimum of 158°K at  $T \approx 25^\circ\text{K}$ .

## I. INTRODUCTION

In general, the specific heats of the rare-earth metals can be expressed as a sum of electronic  $C_E$ , lattice  $C_L$ , nuclear  $C_N$ , and magnetic  $C_M$  contributions.  $C_N$  is due to the splitting of the hyperfine levels by the high magnetic field caused mainly by the orbital angular momentum of 4f electrons of the same atom.  $C_M$  in the rare-earth metals is due to an exchange interaction between unpaired 4f electronic spins of neighboring atoms.  $C_N$  and  $C_M$  are not expected to contribute to the heat capacity of lutetium above 3°K, since in this metal the 4f shell is filled; there are thus no magnetic electrons.

The specific heat  $C_p$  of lutetium has been measured previously between 0.3 and 4°K by Lounasmaa<sup>1</sup> and between 15 and 350°K by Jennings, Miller, and Speeding.<sup>2</sup> In order to fill the gap between these two experiments, we have investigated this metal between 3°K and 25°K. This experiment has also been done to provide data for the analysis of specific-heat measurements on the other trivalent rare-earth metals.<sup>3,4</sup> In order to evaluate  $C_M$  one must subtract from the observed  $C_p$  the lattice and electronic contributions. The coefficient  $\gamma$  in  $C_E = \gamma T$  can be evaluated either from the low-temperature specific-heat data below 4°K for the particular metal or it can be estimated from the values of  $\gamma$  from other rare-earth metals, especially the heavier members of the group from gadolinium to lutetium. Most of these metals have the same outer electronic configuration and the same hcp crystal structure in which the  $c:a$  ratio varies from 1.570 to 1.590. It would not be unreasonable then to take the average  $\gamma$  for the nonmagnetic trivalent lanthanides as a good approximation of the  $\gamma$  for the magnetic lanthanides.

The evaluation of  $C_L$  is not so simple. The low temperature Debye approximation,  $C_L = \alpha T^3$ , is not valid for temperatures above 4°K for the rare-earth metals.  $\Theta_D$  could be calculated if the elastic constants are known at these temperatures, but unfortunately they have not been measured. An indirect approach, which has been used,<sup>4</sup> is to measure  $\Theta_D(T)$  for lutetium.

\* Based on work performed under the auspices of the U. S. Atomic Energy Commission.

<sup>1</sup> O. V. Lounasmaa, Phys. Rev. **133**, A219 (1964).

<sup>2</sup> L. D. Jennings, R. E. Miller, and F. H. Speeding, J. Chem. Phys. **33**, 1849 (1960).

<sup>3</sup> O. V. Lounasmaa, Phys. Rev. **143**, 399 (1966).

<sup>4</sup> O. V. Lounasmaa and L. J. Sundström, Phys. Rev. **150**, 399 (1966).

It is then assumed that  $\Theta_D$  is the same for the heavier rare earths (from Gd to Tm), which were investigated by Lounasmaa and Sundstrom.<sup>4</sup> The assumption for the heavier trivalent rare earths is fairly good. As mentioned above, they all have the hcp structure with similar  $c:a$  ratios. The assumption is also upheld by the observed  $\Theta_D(0^\circ\text{K}) = 195^\circ\text{K}$  for gadolinium and  $\Theta_D(0^\circ\text{K}) = 207^\circ\text{K}$  for dysprosium, which are within the experimental error assigned to the  $\Theta_D(0^\circ\text{K})$  of lutetium. It is the purpose of this research to provide  $\Theta_D(T)$  for lutetium so that such a scheme as just described can be carried out to calculate  $C_L$  for the heavier trivalent rare-earth metals.<sup>4</sup>

## II. EXPERIMENTAL

The specific-heat measurements were made in a cryostat previously described by Lounasmaa.<sup>3</sup> The temperatures were measured with a germanium-resistance thermometer. The original calibration<sup>3</sup> was checked against the vapor pressure of He<sup>4</sup> at two points between 3 and 4°K. This check agreed with the original calibration within 0.5 mdeg K.

The lutetium sample was the same as used by Lounasmaa<sup>1</sup> for his measurements below 4°K. Micro-

TABLE I. The specific heat of lutetium metal (mJ/mole °K). Experimental results (corrected for Ta and Lu<sub>2</sub>O<sub>3</sub> impurities).

$T$	$C_p$	$T$	$C_p$	$T$	$C_p$
3.0114	39.30	5.6956	112.09	10.6422	587.28
3.0151	39.60	5.8291	117.56	10.7539	604.72
3.1356	41.48	5.8394	117.92	11.9347	836.04
3.1782	42.12	6.1410	130.76	12.0991	870.71
3.2428	43.51	6.1543	132.25	12.3188	921.19
3.2788	44.10	6.4176	145.06	13.0856	1113.1
3.4021	46.43	6.5237	150.31	13.7689	1307.9
3.5260	49.15	6.9457	174.48	14.2575	1442.8
3.6872	52.22	6.9787	176.96	14.6402	1565.8
3.7754	54.06	7.1831	189.65	15.5251	1859.5
3.8705	57.48	7.3207	199.53	16.3829	2179.2
4.0307	60.64	7.7502	230.21	17.2326	2499.6
4.0530	60.52	7.8549	238.74	17.5265	2619.1
4.2636	65.65	8.1962	268.54	18.1841	2897.8
4.3908	68.86	8.4170	286.03	19.6495	3533.2
4.5454	72.94	8.5355	299.85	20.0741	3725.3
4.7544	79.19	8.9719	342.12	20.1711	3769.7
4.9232	79.19	9.1145	360.53	21.5527	4422.6
4.9791	87.24	9.2809	382.34	21.8909	4595.2
4.9857	87.48	9.8287	461.36	22.3316	4807.3
5.2994	96.22	9.8326	454.99	23.9315	5629.9
5.3511	98.78	10.1085	500.78	24.7132	6065.3

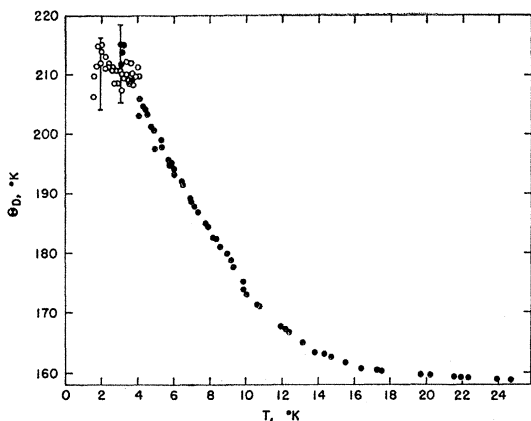


FIG. 1. The Debye temperature  $\Theta_D$  of lutetium metal.

photographs of the sample showed that two minority phases existed in the lutetium sample. Chemically, 0.17% oxygen was found in the sample and 1.4% tantalum. Thus, one of the phases was taken to be  $\text{Lu}_2\text{O}_3$ , so that 1.3% of the lutetium atoms were present as the sesquioxide. The other phase was presumed to be tantalum, since it has been found to precipitate almost completely from the lutetium lattice.<sup>2</sup> The heat capacity is corrected for the impurities by subtracting the impurity heat capacity from the total. The specific heat of tantalum is taken from a compendium of specific-heat data.<sup>5</sup> The specific heat of  $\text{Lu}_2\text{O}_3$  was obtained from unpublished data by Westrum.<sup>6</sup> The final  $C_p$  values were corrected for curvature due to the finite temperature changes when the sample was heated.

### III. RESULTS

The final corrected specific-heat values from our experiment are shown in Table I. Our specific-heat data

<sup>5</sup> R. J. Corroccini and J. J. Gniewek, Nat. Bur. Std. Monograph 21 (1960).

<sup>6</sup> E. F. Westrum (private communication).

agree with Lounasmaa's<sup>1</sup> below 4°K to within 0.6%, and with the smoothed values of Jennings, Miller, and Spedding<sup>2</sup> at 15 and 20°K to within 0.8%. The Debye temperatures were calculated for each data point from Beattie's table<sup>7</sup> by assuming  $C_E=11.27$  mJ/mole°K<sup>1</sup> and  $C_M=C_N=0$ . The values of  $\Theta_D$  calculated are shown in Fig. 1 as the solid circles. The values for  $\Theta_D$  calculated from Lounasmaa's low-temperature data<sup>1</sup> are shown as the open circles in Fig. 1. An error bar for each set of data, representing approximately 95% confidence limits, is also shown in Fig. 1. The bar at lower temperatures refers to Lounasmaa's data and was calculated from the standard deviation of the calculated  $\Theta_D$ 's from 210°K, Lounasmaa's value for  $\Theta_D$  at 0°K. The other error bar refers to our data and was calculated from the standard deviation of the experimental  $C_p$  from a smoothly fitting curve.

The scatter in  $\Theta_D$  at low temperatures is a result of the smallness of the  $C_L$  contribution to the total specific heat; i.e., a  $\frac{1}{2}\%$  statistical error in the total appears as more than a 3% statistical error at 3°K. The scatter becomes so great below 1.5°K that the values of  $\Theta_D$  are meaningless. Our data are in agreement with Lounasmaa's to within the error limits assigned.  $\Theta_D$  drops off from its low-temperature value rather quickly with rising temperature, as is common in hexagonal metals. A minimum of  $\Theta_D=158^\circ\text{K}$  occurs at approximately  $T=25^\circ\text{K}$  before rising to a value of  $\sim 270^\circ\text{K}$  at  $T=\Theta_D$  (calculated from Jennings *et al.*).<sup>2</sup>

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<sup>7</sup> J. A. Beattie, J. Math. Phys. 6, 1 (1926).