THE NUCLEAR CONTRIBUTION TO THE HEAT CAPACITY OF TERBIUM METAL*

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The low temperature heat capacity of terbium metal has been measured by Kurti and Safrata[1] from 6°K to 0.5°K, and by Stantan, Jennings and Spedding[2] from 4°K to 1.4°K. Both measurements indicated an anomaly above 1.0°K, but of different magnitude and shape. This anomaly may well be associated with sample purity. The specific heat data below 1°K given by Kurti and Safrata showed a nuclear contribution which appeared to obey a T^{-2} law to 0.5°K. The objective of the present experiments, which used two cast samples of Ames Laboratory terbium metal similar to that used by Stanton, et al, was first, to use a high purity sample to verify the results of Kurti and Safrata, and second, to extend these results to a lower temperature in an attempt to observe a departure from the T^{-2} dependence.

An ADL magnetic refrigerator, considerably modified, was used to cool the samples, and this established the lowest available temperature as 0.25°K. The heat reservoir of the magnetic refrigerator (also a piece of terbium metal) was connected by a copper rod to copper wires

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imbedded in a paramagnetic susceptibility thermometer (a sphere of powdered potassium chrome alum saturated with DC 550 silicone oil), which was in turn attached to the sample assembly by means of a superconducting lead switch[3]. This thermal switch, which was operated by a liquid nitrogen-cooled solenoid, connected the susceptibility thermometer to the sample assembly, and enabled measurements to be made without having the susceptibility thermometer in thermal contact with the sample.

The sample assembly consisted of two thin pieces of terbium (0.0063 moles each for most of the runs), bolted and glued with GE 7031 adhesive to both sides of a thin copper strip, a manganin heater with superconducting niobium leads wound non-inductively around the terbium, and similarly glued, and a 470 ohm (nominal) 1/2 watt "Old Speer" resistor mounted on the copper strip. The carbon resistor and the susceptibility thermometer were calibrated between 4°K and 1°K using the T_{55E} scale, and the resistor data were fitted to an equation of the form $T_r = a \ln R (\ln R - b)^{-2}$. The term $\underline{a}$ varied slightly (one to three percent) from one run to the next, whereas $\underline{b}$ was roughly constant. Typical values were $\underline{a} = 0.0955$ and $b = 2.715$. Paramagnetic susceptibility temperatures, T_s, were used to obtain corrections to the extrapolated temperatures, T_r, which were obtained from the above equation. The temperature difference, $T_s - T_r$, was less than 0.02° at all temperatures, but decreased rapidly below 0.5°K. This resulted in a fairly large correction to ΔT_r, the

apparent temperature change for a given heat input, in this region. The carbon resistor temperatures were monitored on a 0-1 mv recording potentiometer which was attached to the galvanometer terminals of a Rubicon Type B potentiometer. The procedure was, in most respects, the same as that used by Gaumer and Heer[4] in measuring the atomic heat of sodium metal.

The heat capacity of the non-terbium part of the sample assembly was estimated as roughly four percent at 1°K, and was of much less significance at lower temperatures. The nuclear contribution, C_N, was obtained by subtracting from the data this correction and the values of Stanton, et al, for the spin wave and electronic terms, while the lattice contribution was assumed to be negligible below 1°K. The results for C_N are shown in Fig. 1, where the high temperature data can be expressed as

$$C_N = A \times R \times 10^{-3}/T^2, \qquad (1)$$

with A = 28 (±1) (dashed line in Fig. 1). There is a small deviation near 1°K which may be due to the tail of the anomaly mentioned above. The experimental error above 0.5°K is estimated at about five percent, while at the lowest temperature it could be as large as 10 percent due to uncertainties in the temperature differences.

Kurti and Safrata reported A = 25 (±1), while the paramagnetic resonance experiments of Baker and Bleaney[5] on terbium ethyl sulfate give A = 28.2 (±0.6). Our experimental data agree within experimental

error with Kurti and Safrata at their lowest temperature, 0.56°K, and also at 1°K if the same spin-wave correction term is used. Thus, the discrepancy in the values of A perhaps can be traced to the fact that they used too high a spin-wave correction, and that they should have observed deviations from equation (1) even at 0.56°K. The agreement between the resonance experiments in the dilute salts and the heat capacity measurements on the pure metal was excellent.

The nuclear specific heat of terbium arises from a splitting of the nuclear spin states into four hyperfine levels. This splitting arises from an effective field, H_{eff}, produced by the electronic spins in the 4f orbitals, interacting with the magnetic moment of the terbium nucleus, which has spin I = 3/2.[5] The nuclear specific heat could be given by a formula of the Schottky type[6] if one assumed no direct nuclear spin-spin interaction and an equal spacing, $\Delta/3$, between the four levels. Thus,

$$C_N = \frac{\Delta^2 R}{9k^2T^2}\left\{\frac{\exp(-\frac{\Delta}{3kT})\left[1+\exp(-\frac{\Delta}{3kT})\right]^4 + 4\exp(-\frac{\Delta}{kT})}{\left[1+\exp(-\frac{\Delta}{3kT}) + \exp(-\frac{2\Delta}{3kT}) + \exp(-\frac{\Delta}{kT})\right]^2}\right\}. \quad (2)$$

The specific heat as calculated from this equation is shown also in Fig. 1 (solid line), with Δ= 0.45°K, corresponding to the high temperature value of A = 28. The disagreement between the calculated curve and the experimental data would seem to be slightly outside the experimental error at the lowest temperatures. Equation (2) would predict a maximum in the specific heat at about 0.1°K, with a value of C_N of the order of

1.5 cal/mol-deg. The value of Δ also allows one to use the magnetic moment of terbium as given by Baker and Bleaney[7] (μ = 1.52 (±0.08) nuclear magnetons) to calculate $H_{eff} = 4 \times 10^6$ gauss.

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FIGURE CAPTIONS

Figure 1. The experimental data compared with Equations (1) and (2) with A=28 in Equation (1).

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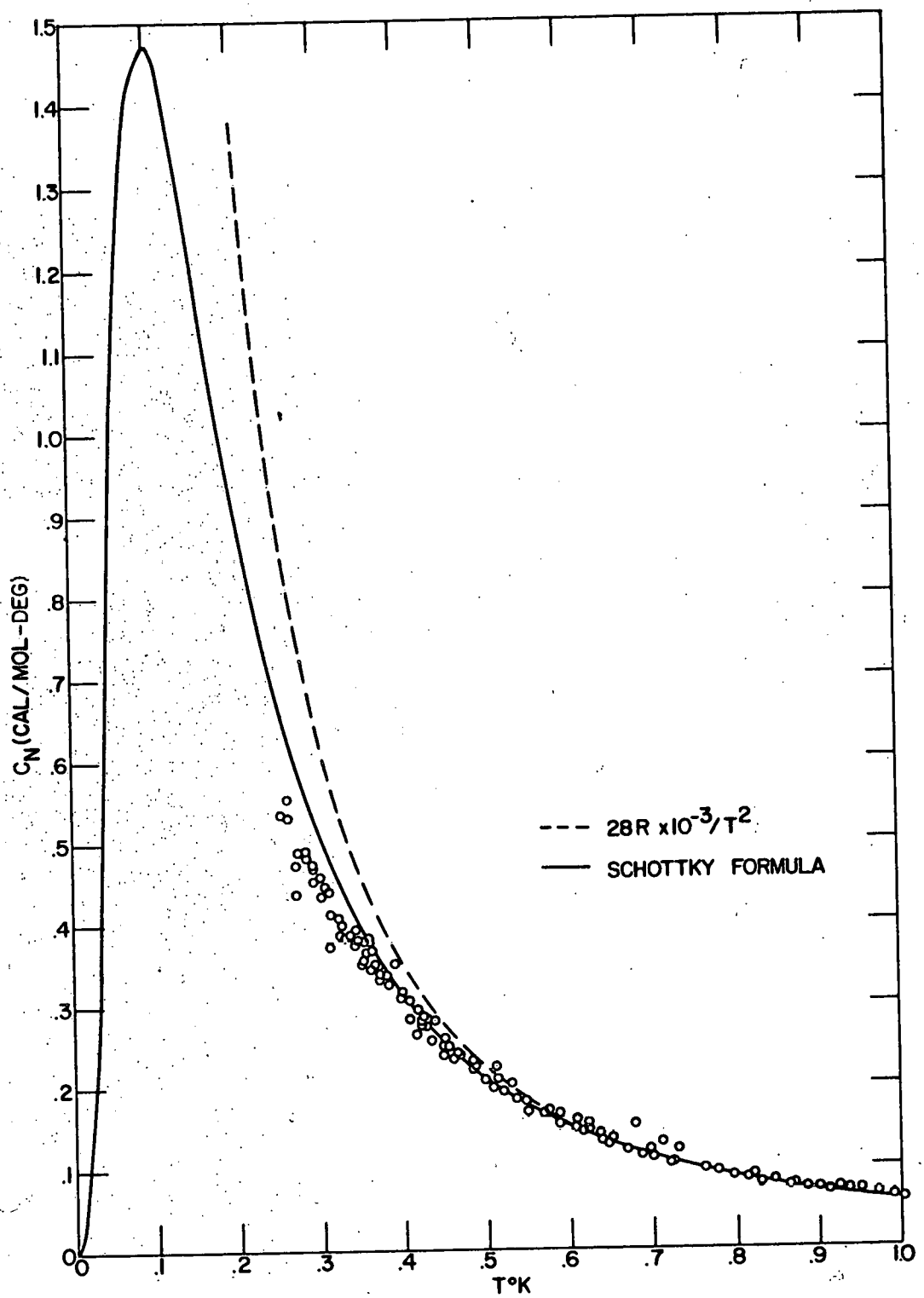
1.5
1.4
1.3
1.2
1.1
1.0
.9
.8
.7
.6
.5
.4
.3
.2
.1
0
C_N (CAL/MOL-DEG)
0
.1
.2
.3
.4
.5
.6
.7
.8
.9
1.0
T°K
--- 28R x10^{-3}/T^2
—— SCHOTTKY FORMULA

Figure 1.