

MAGNETIC ORIGIN OF THE OBSERVED ANOMALY IN THE SPECIFIC HEAT OF GADOLINIUM AT 3.8 K¹⁾

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The specific heat of gadolinium has been measured between 2.5 K and 10 K using an adiabatic pulse method. The anomaly at 3.8 K is associated with the magnetic ordering of a Gd₂O₃ impurity. Owing to the magnetic field of 3 T this anomaly decreased and shifted to a lower temperature.

The electronic specific heat constant $\gamma = 19.54 \pm 1.12$ mJ/mol K² and the low temperature limiting Debye temperature was 184.8 ± 0.3 K. A magnetic contribution was not observed.

МАГНИТНАЯ ПРИРОДА НАБЛЮДАЕМОЙ АНОМАЛИИ В УДЕЛЬНОЙ ТЕПЛОЕМКОСТИ ГАДОЛИНИЯ ПРИ ТЕМПЕРАТУРЕ 3,8 К

В работе при помощи адиабатического импульсного метода измерена температурная зависимость удельной теплоемкости поликристаллического гадолиния в диапазоне температур 2,5—10 К в магнитных полях с индукцией до 3Т. В диапазоне температур 3,7—3,8 К обнаружена аномалия в поведении теплоемкости, которая обусловлена упорядочением в магнитной примеси Gd₂O₃. В магнитных полях с индукцией до 3Т эта аномалия сильно подавлена. Низкотемпературное ограничение на температуру Дебая равно $184,8 \pm 0,3$ К, а электронная удельная теплоемкость равна $\gamma = 19,54 \pm 1,12$ мДж/моль·К². Не обнаружен магнитный вклад в удельную теплоемкость.

1. INTRODUCTION

The specific heat C_p of heavy rare earths at low temperatures consists of four parts, that is C_L — lattice contribution, C_E — electronic contribution, C_M — magnetic contribution and C_N — nuclear contribution

$$C_p = C_E + C_L + C_M + C_N \quad (1)$$

where $C_E = \gamma T$ and $C_L = \beta T^3$.

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According to [1] the contribution of C_v in the specific heat of gadolinium is not present in the temperature range from 1 K to 10 K.

The measured values of the specific heat of Gd are given in [2]. The authors of this work supposed that the apparent anomaly found between 3 K and 5 K is caused by impurities consisting mainly of Gd_2O_3 . The magnetic contribution has the value $C_m \sim T^{2.7}$. This anomaly was not found in later measurement on the Gd sample purified by SSE method [3]. From the value obtained on such purified sample [3] the value of electronic contribution $\gamma = 3.7$ mJ/mol K² and that of the Debye temperature $\Theta_D(0) = 187 \pm 3$ K was determined, the latter being in good agreement with the value $\Theta_D(0) = 184$ K determined from the measurements of elastic constants [4]. The magnetic contribution was found to be $C_m \sim T^{1.75}$.

In [5] the results obtained on the Gd sample with RRR = 66 are given. From this measurement the value $\gamma = 8.2$ mJ/mol K² was determined. After purifying the sample by the SSE method and subsequent annealing in the ultrahigh vacuum the value of γ changed by 20%. From these measurements the Debye temperature $\Theta_D(0) = 184$ K and the value $\gamma = 6.5$ mJ/mol K² were determined. No magnetic contribution to specific heat was observed.

II. RESULTS AND DISCUSSION

The measurements were made on a cylindrical Gd sample with a diameter of 4.8 mm and a mass of 15.22×10^{-3} kg. The purity as quoted by Metalimex, USSR, was 99.8%, the main impurities being oxygen (0.1%) and rare earth (0.1%). Our analysis by means of an Exhalograf Baltzer showed the presence of oxygen (0.2813%) and hydrogen (30 ppm). Other elements were not determined. The analysis showed that oxygen diffused mainly to the surface layers of Gd.

The method of specific heat measurements is described in [6] and the apparatus in [7]. The accuracy of the measurements was better than $\pm 1\%$ in the whole range from 2 K to 10 K.

Fig. 1 shows the results of the measurement of the Gd specific heat in the temperature range from 2 K to 10 K. Anomalies were observed on the $C(T)$ dependence — one at temperatures between 3.7 K and 3.8 K and another at 3 K. According to the chemical analysis of the sample and using the results of [8] the specific heat of Gd_2O_3 was separated in our measurements. This separation showed a higher concentration of oxygen in the sample than that determined by chemical analysis. After evaluating the ratio of the surface S_1 (for the analysed sample) to the original surface S_2 we concluded that the oxygen concentration in our sample was 0.3%. The result of the separation of the specific heat of Gd_2O_3 for the oxygen concentration of 0.3% is given in Fig. 2.

The observed values were fitted — using the least squares method — by the polynomial.

$$C_p = \gamma T + \beta T^3 \quad (2)$$

and from this the values of $\gamma = 19.54 \pm 1.12$ mJ/mol K² and $\Theta_D(0) = 184.8 \pm 0.3$ K were determined.

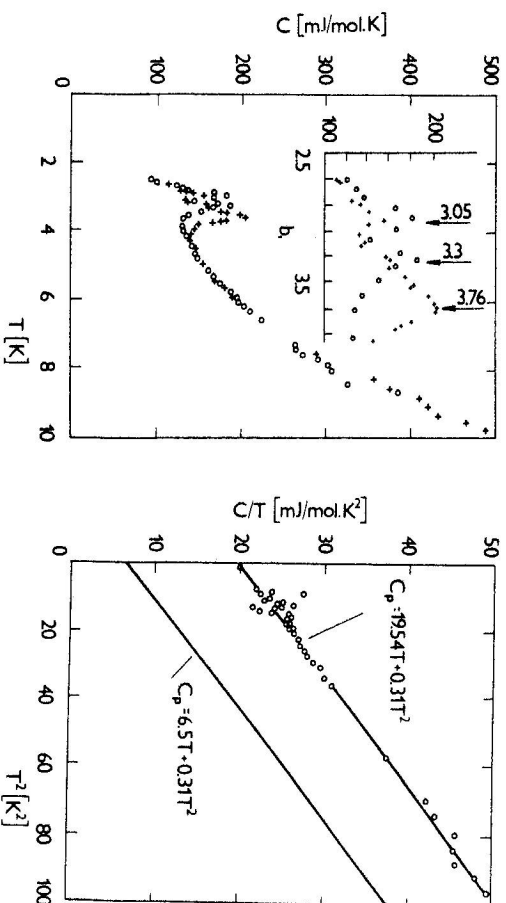


Fig. 1. Specific heat of gadolinium in the magnetic field 3 T (□) and without the magnetic field (+).

Fig. 2. Specific heat of gadolinium after separation of the specific heat of Gd_2O_3 .

The fitted dependence is shown in Fig. 2 together with the fitting polynomial according to [5]. The determined value of the Debye temperature is in very good agreement with the value calculated from the elastic constants [6] and from the specific heat [5, 3].

The apparent difference in the values of γ may be probably explained either by the presence of impurities in our sample or by a higher density of states on the Fermi surface. This should be proved by further measurements.

It seems that the value of the magnetic contribution has no statistical importance in the temperature range from 2.5 K to 10 K, hence we conclude that — in agreement with [5] — $C_m = 0$.

The measurement of the specific heat of gadolinium in the magnetic field of 3 T provided an experimental evidence of the presence of a magnetic impurity at a temperature of 3.7 K. The magnetic field suppresses strongly the anomaly caused

by Gd_2O_3 , lowering both its absolute value and the transition temperature (Fig. 1b). The anomaly at 3 K should be explained by further measurements.

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