latorische Korrelationszeit von hohen Temperaturen her extrapoliert. Die durchgezeichneten Kurvenzüge berücksichtigen einen Rotationsanteil mit $K_r/K_t = 0{,}3$ und den in Tab. 1 gegebenen Werten für $\tau_r$ und $E_r$. Es ist dies als eine Bestätigung der in Anm. [4] gegebenen Interpretation aufzufassen.

### Schlußfolgerungen

Die Messungen der Spin-Gitter-Relaxationszeiten bestätigen im wesentlichen die aus Untersuchungen der dynamischen Kernpolarisation gewonnenen Kenntnisse über Lösungen freier Radikale. Sie zeigen, daß der translatorische Diffusionsmechanismus vorherrscht. Eine Ausnahme bilden die wäßrigen Lösungen des $(SO_3)_2NO^{--}$-Ions. Nur die Ergebnisse an $C_6F_6$ machen weitere Messungen an fluorierten Lösungen und ein verfeinertes Modell erforderlich. Die Untersuchungen haben ferner gezeigt, daß sich die Korrelationszeiten in solchen Systemen überraschend gut mittels der STOKES–EINSTEIN-Beziehung aus der Viskosität berechnen lassen, wenn man nur die Lösungsmittelmoleküle berücksichtigt, und für diese gemittelte Molekülradien einsetzt.

# Electrical Resistivity, Thermal Conductivity and Magnetic Susceptibility of Polycrystalline Samarium at Low Temperatures

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Electrical resistivity, thermal conductivity, and magnetic susceptibility have been measured, using the same sample of samarium, from 4 to 300 °K, from 5 to 200 °K, and from 4 to 300 °K, respectively. Two anomalies, one at $12 \pm 1$ °K and another at $106 \pm 1$ °K, are observed, resulting from an order-order magnetic transformation and an antiferromagnetic-paramagnetic transition, respectively. The LORENZ function is found to be larger at any temperature than that expected for pure electronic thermal conductivity. This implies that there is some phonon and possibly also some magnon thermal conductivity in samarium at low temperatures. The magnetic moment disorder electrical resistivity of samarium is determined to be $39.0 \pm 0.5$ $\mu\Omega$ cm, in fair agreement with the value to be expected from theoretical considerations.

According to studies of the magnetic susceptibility [1–3], specific heat [4, 5], electrical resistivity [6–8], and thermoelectric power [9], polycrystalline samarium exhibits two magnetic transitions at low temperatures. One occurs at about $12 \pm 1$ °K and the other at $106 \pm 1$ °K. Neutron diffraction investigations have not yet been done on samarium. Thus, the nature of these transitions is not clearly understood at the present time. However, on the basis of the behavior of the above-mentioned physical properties, it appears that the transition at 106 °K is the NÉEL point, i.e., a transition from an antiferromagnetic state to the paramagnetic state. The low temperature transition is quite likely a magnetic order-order transition. Recently we have found [10–13] that the changes in the magnetic state in gadolinium, dysprosium, terbium, and erbium, whose detailed moment arrangements have been determined by neutron diffraction techni-

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ques [14–17], exhibits noticeable influence on the heat conductivity of these metals. Since thermal transport studies have not yet been done on the light rare earth metals, we decided to perform such measurements. In this paper we present the results of electrical resistivity, thermal conductivity, and magnetic susceptibility of samarium and discuss their significance.

## Experimental Considerations

The sample of samarium, whose electrical resistivity, thermal conductivity, and magnetic susceptibility were measured as a function of temperature, was prepared in the following manner. The initial stock of distilled samarium was obtained from Research Chemicals. According to the supplier, a partial analysis showed the following impurities in weight %: Eu – 0.05, Ca – 0.02, Mg – 0.01, Gd – 0.01, and Si – 0.005. The metal was arc melted for 10 minutes in 100 Torr argon atmosphere. After the melting, a rod of 0.479 cm diameter and about 6 cm length was machined from the ingot. Swaging of samarium ingots resulted in fracture, and it was therefore impossible to use this method to produce a good polycrystalline structure. The electrical resistivity of this sample at 4.17 °K was found to be 6.73 $\mu\Omega$ cm. The electrical resistivity and thermal conductivity data were obtained using the equipment described elsewhere [18, 19]. Magnetic susceptibility measurements were made using a force method [20]. The sample for the susceptibility studies was a section cut from the transport property specimen.

## Results and Discussion

The electrical resistivity of samarium from liquid helium temperatures to 300 °K is shown in Fig. 1. One clearly can observe two anomalies at low temperatures: one at 12 °K and the other at 106 °K. The detailed temperature variation of the electrical resistivity in the neighborhood of these temperatures is shown in Figs. 2 and 3, respectively. Since the

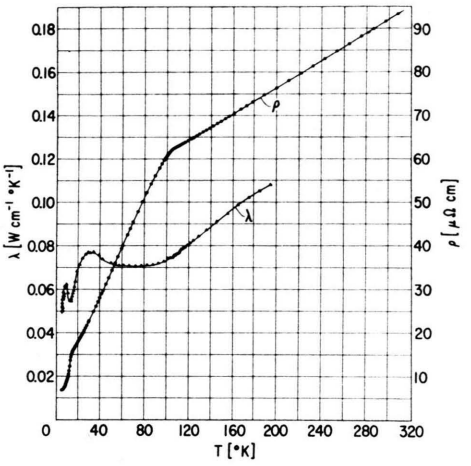


Fig. 1. Electrical resistivity and thermal conductivity of samarium as a function of temperature.

temperature coefficient of the electrical resistivity, defined by

$$\alpha = (1/\varrho)\,(\mathrm{d}\varrho/\mathrm{d}T), \qquad (1)$$

where $\varrho$ is the total electrical resistivity at a temperature $T$, is a particularly sensitive function for detecting the magnetic transitions, we also have plotted this quantity in Figs. 2 and 3. From these plots the above-given transition temperatures were determined.

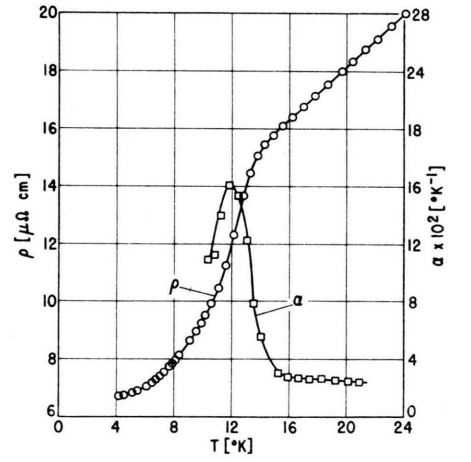


Fig. 2. Electrical resistivity of samarium and its temperature coefficient in the neighborhood of 12 °K.

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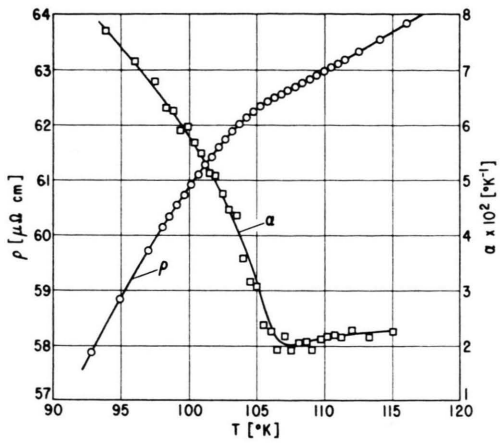


Fig. 3. Electrical resistivity of samarium and its temperature coefficient in the neighborhood of 106 °K.

The electrical resistivity of samarium changes almost linearly with temperature below 300 °K. At about 106 °K the resistivity decreases much more rapidly with decreasing temperatures than above this temperature. This behavior strongly suggests that above 106 °K the magnetic moments, resulting from the localized 4 f electrons in samarium, are disordered and give rise to considerable moment-disorder resistivity [21, 22]. We estimate this from the experimental data to be $39.0 \pm 0.5\ \mu\Omega$ cm. If one neglects the transfer of energy between the conduction electrons and the magnetic moment system, the moment-disorder electrical resistivity is given by [23, 24]

$$\varrho_{\mathrm{md}} = \frac{3\pi N m^*}{2\hbar e^2 E_{\mathrm{F}}} G^2 (g-1)^2 J(J+1), \qquad (2)$$

where $N$ is the number of atoms per unit volume, $m^*$ the effective of a conduction electron, $e$ the electronic charge, $E_{\mathrm{F}}$ the FERMI energy, $g$ the LANDÉ $g$-factor, $J$ the quantum number associated with the total angular momentum of the magnetic moment, and $\hbar = h/2\pi$, $h$ being the PLANCK constant. The symbol $G$ in Eq. (2) represents a coupling constant with the dimensions of energy times volume. Since the quantity

$$[(3\pi N m^*)/(2\hbar e^2 E_{\mathrm{F}})]\, G^2$$

in Eq. (2) is expected to be approximately the same for all trivalent rare earth metals, one predicts that $\varrho_{\mathrm{md}}$ should be proportional to $(g-1)^2 J(J+1)$. Since this factor is 4.46 and 4.50 for samarium and holmium, respectively, resistivity should be approximately equal for these two rare earth metals. This, indeed, is in accordance with the experimental results; for holmium [22] $\varrho_{\mathrm{md}} = 32.3\ \mu\Omega$-cm which should be compared with $\varrho_{\mathrm{md}} = 39.0\ \mu\Omega$ cm for samarium.

The thermal conductivity of samarium, also shown in Fig. 1, exhibits two maxima at low temperatures, one at about 9 °K and another at 32 °K. The first maximum and the following minimum at 13 °K result from the low temperature magnetic order-order transformation. The observed thermal conductivity behavior is qualitatively consistent with the electrical resistivity behavior, i.e., the rapid increase in the resistivity above 10 °K causes a decrease in the thermal conductivity. The second maximum is, of course, of the conventional type observed in all pure metals. This maximum, qualitatively can be understood if one recalls that at low temperatures ($T \ll \Theta$, where $\Theta$ is the DEBYE temperature) the electronic thermal conductivity, limited by the impurity scattering, is proportional to $T$. At higher temperatures ($T \approx \Theta$) the electron-phonon scattering causes the electronic thermal conductivity to follow approximately $T^{-2}$ dependence. Thus, a maximum in the $\lambda$ vs. $T$ curve results.

The thermal conductivity change at the NÉEL point, which according to the electrical resistivity data is 106 °K, is not abrupt but gradual. However, the general increase in the thermal conductivity, when the magnetic moment become discordered, is in agreement with the results on the other rare earth metals [10, 12, 13] above their NÉEL temperatures.

In principle, the total thermal conductivity of a rare earth metal, possessing an ordered magnetic structure, should be written as

$$\lambda = \lambda^{\mathrm{e}} + \lambda^{\mathrm{l}} + \lambda^{\mathrm{m}}, \qquad (3)$$

where the superscripts, e, l, and m imply that the heat is carried by electrons, phonons, and magnons, respectively. For pure electronic thermal conductivity at low temperatures ($T \ll \Theta$), where the scattering processes can be considered elastic, one would expect that the LORENZ function

$$L = \varrho\, \lambda / T \qquad (4)$$

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should be equal to the LORENZ number

$$L_0 = \pi^2 k^2/3\, e^2 = 2.445 \times 10^{-8}\ [\mathrm{V^2\,{}^\circ K^{-2}}]. \quad (5)$$

In this equation $k$ is the BOLTZMANN constant and $e$ the electronic charge. If some heat is also transported by phonons and magnons, then $L$ should be larger than $L_0$. Calculations of the LORENZ function from the experimental data clearly establish that the experimental value of $L$ is considerably larger than $L_0$ at any temperature. This implies that there is some heat conduction by phonons and also possibly magnons. This aspect of the quantity $L$ for samarium is similar to the behavior found in dsyprosium [10], gadolinium [11], terbium [12], and erbium [13]. Since the present theoretical understanding of the role of magnons in the total heat conductivity of a magnetically ordered metal is quite poor, it is difficult at this moment to separate the term $\lambda^l$ and $\lambda^m$ from the measured values of $\lambda$. We are planning in the near future to perform thermal conductivity measurements on the above-mentioned rare earth metals (and also other ferromagnets) in the presence of strong magnetic fields (up to 60 kOe) in order to learn more about the magnitude and temperature dependence of $\lambda^m$.

In regard to the thermal conductivity studies, we note that recently accurate thermal conductivity measurements on rare earth metals at 291 °K have been made by POWELL and JOLLIFFE [25]. For samarium they find $\lambda = 0.133\ \mathrm{W\,cm^{-1}\,{}^\circ K^{-1}}$ and $L = 4.2 \times 10^{-8}\ \mathrm{V^2\,{}^\circ K^{-2}}$. These values appear to be consistent with our measurements when extrapolated from 200 °K to 291 °K. We did not perform thermal conductivity studies on samarium above 200 °K because of serious difficulties experienced with radiation corrections. For this reason our previous data on other rare earth metals [10–13] above liquid nitrogen temperatures now appear to be higher than the true values as can be concluded from the work by POWELL and JOLLIFFE.

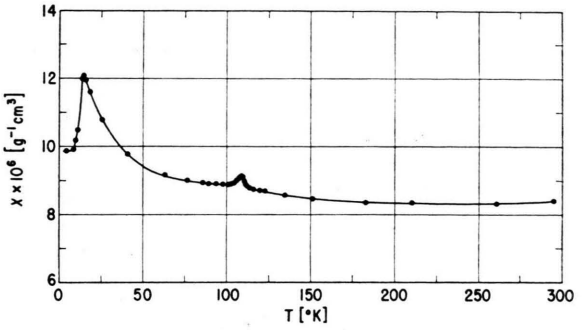


Fig. 4. Magnetic susceptibility of samarium as a function of temperature.

Magnetic susceptibility ($\chi$) of samarium, used in this investigation, is shown in Fig. 4. The data were taken with increasing temperatures, starting at 4.2 °K, and using a magnetic field of 5 kOe. Figure 4 clearly shows two peaks in the $\chi$ vs. $T$ curve: one occurs at 14 °K and the other at 108 °K. Our measurements are in reasonable agreement with those obtained by other investigators [1–3], except that the anomaly at 108 °K appears to be more clearly observable from our data than from the previous studies.

*Acknowledgments*

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