This reduces to Eq. (27) of I if we set $P_v^N=0$, $\eta_{||}=0$, $\eta_\perp=1$. Although we will not calculate it, the nuclear polarization also affects the final neutron polarization in all three peaks.

It may be in order to point out several problems involved in performing the experiments we have indicated. In a typical sample, diffraction occurs from many "mosaic blocks," rather than from a single large crystallite. A portion of the beam diffracted by the upper layers of the crystal may be rediffracted into the incident direction by deeper layers; this is, in fact, part of the familiar phenomenon of secondary extinction. Since the polarization of the diffracted beam may be changed in magnitude and direction, the analysis of the rediffraction process may be extremely complicated, and it is thus important to ensure that the experimental crystal is extinction free.

While the above diffraction process occurs only at occasional crystallites, absorption will occur throughout the body of the crystal. Thus, even though the absorption is small enough so that the nuclear amplitudes in (20) and (21) may be taken as real, absorption may still play a significant role in the experiment. The absorption is, in general, spin-dependent—that is, it is different for the $I+\frac{1}{2}$ and $I-\frac{1}{2}$ nuclear states. Further, the spin dependence of the absorption cross section may be opposite to that of the scattering, leading to possibly erroneous results. Again, as in secondary extinction, the analysis becomes very complicated if the crystal is too thick. However, if the crystal is extinction-free, then each spin component of the incident beam may be treated individually. The usual thin crystal-diffraction solutions then apply to each component separately, each with its own appropriate absorption coefficient. In principle, this coefficient should include both incoherent scattering and true absorption, but in practice only true absorption fiill be important. The spin dependence of this quantity is given by[8]

$$\sigma_a=\sigma_{0a}+\sigma_{pa}\mathbf{P}^N\cdot\mathbf{P}, \tag{27}$$

where

$$\sigma_{0a}=[(I+1)/(2I+1)]\sigma_a^{+}+[I/(2I+1)]\sigma_a^{-},$$

$$\sigma_{pa}=[I/(2I+1)](\sigma_a^{+}-\sigma_a^{-}),$$

and $\sigma_a^{\pm}$ are the absorption cross sections for the two possible compound states. Although σ_{pa} is unknown for most nuclei, it may be determined by studying the transmission of the incident beam with the crystal rocked out of the reflecting position (see, for instance, Ref. 9).

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Elastic Moduli and Ultrasonic Attenuation of Polycrystalline Europium from 4.2 to 300°K

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The longitudinal and transverse acoustic velocities and the ultrasonic attenuation of high-purity polycrystalline europium metal have been measured by a pulse technique between 4.2 and 300°K. The variations with temperature of the Young modulus E, shear modulus G, adiabatic compressibility K_s, and Debye temperature Θ_D, have been determined. The Néel point at 91°K is marked by drastic changes in the elastic moduli and ultrasonic attenuations. Anomalies in the attenuation and in the compressibility are observed at about 150°K. The limiting value of Θ_D is 117°K.

INTRODUCTION

SEVERAL physical properties of europium differ from those of the other rare-earth metals: for instance, its bcc structure, in contrast to the mostly hexagonal metals of the series, the large atomic volume,[1] and the high compressibility at room temperature.[2]

The magnetic susceptibility[3] exhibits a small antiferromagnetic-type maximum at 90°K. No remanence was observed, i.e., no ferromagnetic state occurs. Nevertheless, europium does not display a typical antiferromagnetic behavior. The high-temperature susceptibility is consistent with the fact that the Eu ions are in a divalent state. At lower temperatures, however, the susceptibility fits a trivalent model.

Neutron diffraction measurements[4] show that Eu undergoes an antiferromagnetic transition, with a Néel point at 91°K, followed by a continuing ordering

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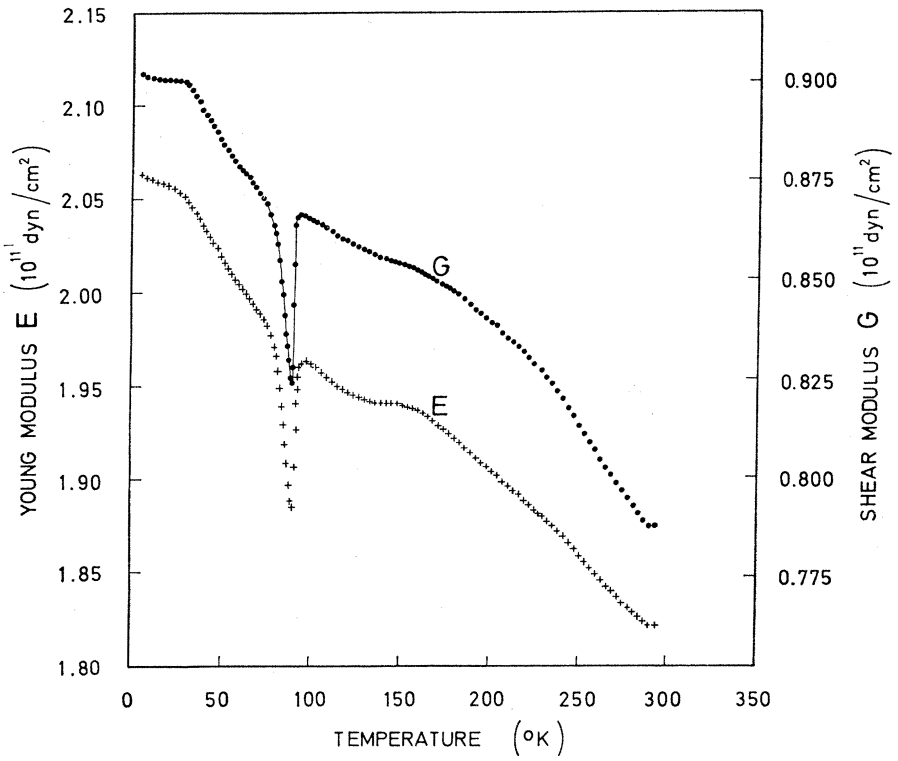

FIG. 1. The temperature variation of the Young (E) and shear moduli (G) of high-purity polycrystalline europium metal.

process until saturation at 20°K. The measured magnetic moment found was 5.9 μ_B, which is between the maximum theoretical values of 7 and 3 μ_B, expected for the di and trivalent Eu ions, respectively. From the neutron diffraction intensity data[4] an attempt was made to calculate the Debye temperature. At the lowest temperature quoted (100°K), $\Theta_D=68$°K. Lounasmaa[5] assumed $\Theta_D=100$°K for the analysis of his low-temperature heat-capacity measurements.

The electrical resistivity[6] displays a sharp peak at 90±3°K, followed by a decrease in the resistivity with increasing temperature. The negative resistivity coefficient persists up to about 145°K, where it becomes positive again. It appears that the minimum in the electrical-resistivity curve corresponds to the point at which the plot of the reciprocal of the magnetic susceptibility versus temperature departs from the Curie-Weiss law.[7]

There are no published data available on the elastic and anelastic behavior of europium below room temperature. The objective of the present work was to study the variation of the elastic moduli and the ultrasonic attenuation from 4.2 to 300°K. A magnetic contribution to the elastic moduli was expected in the vicinity of the magnetic transition.[8] In addition, the variation of the Debye temperature and the adiabatic compressibility with temperature should be useful in the analysis of heat-capacity and high-pressure data.

EXPERIMENTAL

High-purity (99.9+%) europium disks, supplied by Johnson Matthey Co., England, were used. The specimens were 12 mm in diameter, and about 6 mm thick. Europium metal reacts rapidly with moist air. Therefore, special handling techniques were necessary to prevent surface reactions during the preliminary measurements, e.g., density determination and surface preparation of the samples.

An inert-atmosphere enclosure was constructed. The high-purity helium gas pressure inside the enclosure was slightly above the atmospheric. Thus, air leakage into the enclosure was avoided. The room-temperature density was determined by means of a fluid displacement method, using monobromobenzene, and was found to be 5.285 g cm^{-3}. After hand-lapping the sample surfaces to a parallelism of better than 1 part in 10^4 the thickness was measured to within 5×10^{-4} mm. The specimen was then mounted in the holder and rapidly transferred from the inert atmosphere enclosure into the cryostat. No visible reaction was apparent on the sample surfaces after the low-temperature runs.

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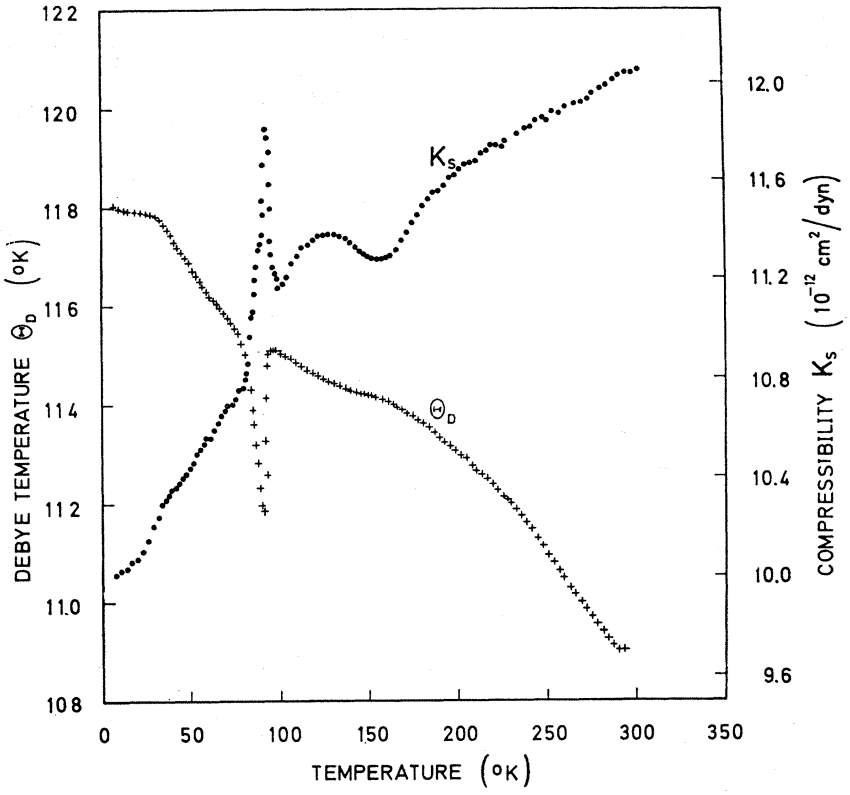

FIG. 2. The temperature variation of the adiabatic compressibility K_s and the Debye temperature Θ_D of high-purity polycrystalline europium metal.

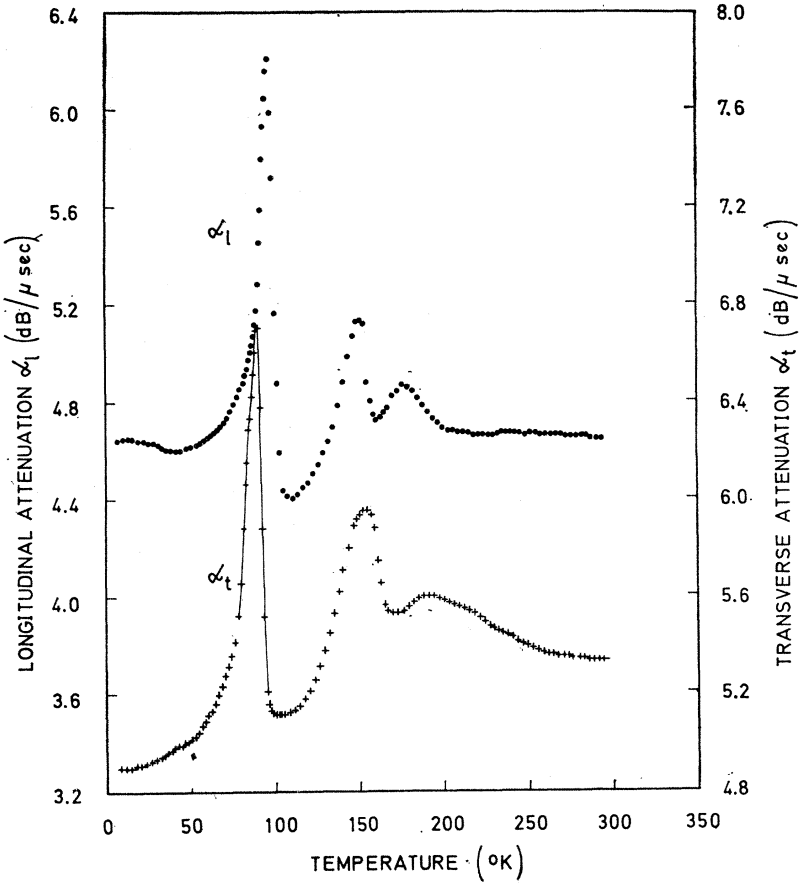

FIG. 3. The temperature variation of the longitudinal (α_l) and transverse (α_t) attenuation of high-purity europium metal.

The acoustical path lengths of the europium specimens were corrected by taking 30×10^{-6} deg^{-1} as the linear coefficient of thermal expansion below room temperature.[9] It is assumed that the coefficient of thermal expansion varies discontinuously in the vicinity of the magnetic transition point. Taking a linear expansion coefficient of 100×10^{-6} deg^{-1} over a range of 10°K leads to an uncertainty in the specimen thickness of 0.1%, which is still within the limits of the experimental error.

The elastic moduli and Debye temperatures were calculated from the experimentally determined longitudinal and transverse sound velocities. A pulse technique was employed at a frequency of 10 MHz. Experimental details and method of data analysis will be described elsewhere.[8] The estimated experimental error in the absolute values of the elastic moduli is 0.4%. The relative, point to point, precision is better by a factor of 4. The ultrasonic attenuation was determined to within 1%, and the temperature to 0.5°K.

RESULTS AND DISCUSSION

The temperature dependence of the elastic moduli of polycrystalline Eu is given in Fig. 1. The symbols E and G represent the Young and shear moduli, respectively. Both E and G display sharp minima at the Néel point of 91°K. Such a behavior is consistent with a typical antiferromagnetic transition. A corresponding λ-type anomaly is observed in the adiabatic compressibility K_s (Fig. 2). This is qualitatively in agreement with the theory of second-order phase transformations of Landau and Lifshitz.[10]

The critical fluctuations of the spins in the vicinity of a magnetic ordering point interact with the acoustical phonons, giving rise to anomalies in the ultrasonic attenuation. This behavior was predicted by the Landau-Khalatnikov theory.[11] According to this theory, the ultrasonic attenuation should display a peak on the ordered (low-temperature) side of the magnetic transition point. Both longitudinal and shear wave attenuations (Fig. 3) exhibit such anomalies. The peak in the attenuation occurs approximately 5°K below the Néel point, compared to 32°K in α-Mn.[12] The temperature of the attenuation maximum can be related to the ultrasonic frequency employed and to a relaxation time which characterizes the phenomenon. The kinetics can be established if relaxation times are determined at several frequencies.

The ultrasonic attenuations (Fig. 3) and the compressibility (Fig. 2) show anomalies in the vicinity of 150°K. In contrast to the anomalies in the elastic moduli and attenuations at 91°K, which are connected to the antiferromagnetic ordering, the ones of 150°K do not exhibit the typical λ character. The Young and shear moduli, and consequently, the compressibility, are affected to a limited extent only. The attenuations, on the other hand, show well-defined peaks. This behavior is related to the occurrence of a minimum in the electrical resistivity[6] and a change in slope of the reciprocal value of the magnetic susceptibility versus temperature.[7] The observed anomalies in the ultrasonic attenuations are probably connected to the negative temperature coefficient of the electrical resistivity between 150°K and the Néel point.[6] This behavior is apparently due to a change in the electronic structure of europium metal, caused probably by an interband electron transfer mechanism.

Figure 2 shows the temperature dependence of the Debye temperature. Since the Debye temperature is calculated from sound velocities, its variation with temperature should be similar to that of the elastic moduli. A sharp drop is observed at the Néel point. According the the 3rd law of thermodynamics, the elastic moduli and Debye temperature should approach absolute zero with zero slope. Such a behavior was observed in the present study. The limiting Debye temperature $\Theta_D{}^0$, extrapolated to 0°K, was found to be 117°K. The room-temperature value of Θ_D was 110°K.

Low-temperature heat-capacity measurements[5] revealed a peak at 16°K. It was attributed to the occurrence of a cooperative phenomenon. Although the elastic moduli, and particularly, the ultrasonic attenuations, are sensitive to magnetic ordering phenomena, no such anomaly has been found in the course of the present work.

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