

Seebeck Coefficients of Lutetium Single Crystals*

L. R. EDWARDS,† J. SCHAEFER,‡ AND S. LEGVOLD

Institute for Atomic Research and Department of Physics, Iowa State University, Ames, Iowa 50010

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The Seebeck coefficients of lutetium have been measured along the $[10\bar{1}0]$ (b axis) and $[0001]$ (c axis) crystallographic directions as a function of temperature from 8 to 300 K. These results are compared with the single-crystal Seebeck coefficients of yttrium and discussed in terms of the Fermi surface.

SINGLE-CRYSTAL Seebeck coefficients of the heavy rare earths (Gd, Tb, Dy, Ho, Er, and Tm) have been measured and found to be very anomalous and anisotropic.^{1,2} The anomalous behavior in these materials is due to magnetic interactions, while the anisotropic behavior is probably due to the anisotropy of the Fermi surface. Lutetium is the last element in the rare-earth series and has a full complement of electrons in the $4f$ shell (14). Thus in lutetium there will be no magnetic interaction and magnon drag effects. The Seebeck coefficients will have only diffusion and phonon drag components. If a theoretical understanding of the Seebeck coefficients of lutetium could be accomplished, then some insight might be gained into the complex Seebeck coefficients of the magnetic rare earths.

Yttrium and lutetium have very similar electronic structures (similar Fermi surfaces) and should therefore have similar transport properties. Here we report the Seebeck coefficients of lutetium single crystals and compare the results with the Seebeck coefficients of yttrium.

The lutetium single crystals used in this study were the same as those used by Boys and Legvold³ for thermal conductivity measurements. Two samples with dimensions $2 \times 2 \times 10$ mm were used: one with the $[0001]$ direction (c axis) along the sample axis and the other with the $[10\bar{1}0]$ direction (b axis) along the sample axis. Sample purity and method of preparation are given in Ref. 3, and the experimental apparatus is described in Ref. 2.

The Seebeck coefficients of lutetium were measured as a function of temperature from 8 to 300 K; the results are shown in Fig. 1. For comparison, the single-crystal Seebeck coefficients of yttrium¹ are also shown in Fig. 1. For lutetium above 70 K, the c -axis Seebeck coefficient increases with increasing temperature and goes positive, while the b -axis Seebeck coefficient is fairly independent of temperature and remains negative. The Seebeck coefficients of lutetium exhibit considerable anisotropy as compared to yttrium.

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† Present address: Sandia Laboratories, Albuquerque, N. M. 87115.

‡ Present address: Northwestern University, Evanston, Ill. 60201.

¹ L. R. Sill and S. Legvold, Phys. Rev. **137**, 1139 (1965).

² L. Roger Edwards and S. Legvold, Phys. Rev. **176**, 753 (1968).

³ D. W. Boys and S. Legvold, Phys. Rev. **174**, 377 (1968).

The Seebeck coefficient of a nonmagnetic material arises from electron and hole diffusion and phonon drag (both normal and umklapp processes). To a first approximation these contributions are additive

$$S = S_e + S_p. \quad (1)$$

For normal processes, a phonon drag peak should be observed in the range $\theta_D/10 - \theta_D/5$. For lutetium this should be around 20 K; however, experimentally a peak is observed around 70 K.

According to Ziman,⁴ the diffusion Seebeck coefficient can be expressed by

$$S_i = \frac{\pi^2 k^2 T}{3e} \left(\frac{\partial \ln \sigma_i(\epsilon)}{\partial \epsilon} \right)_{\epsilon_F} \quad (2)$$

and

$$\sigma_i(\epsilon) = \frac{e^3}{4\pi^3 \hbar} \int_{\epsilon} \tau v_i dS_i, \quad (3)$$

where τ is the relaxation time, v_i is the electronic velocity in the i th direction on a surface of constant energy ϵ , and dS_i is an area element on the energy surface ϵ . In the case of cubic symmetry, Eqs. (2) and (3) become

$$\sigma = (e^2/4\pi^3 \hbar) \Lambda S \quad (4)$$

and

$$S = \frac{\pi^2 k^2 T}{3e} \left(\frac{\partial \ln \Lambda}{\partial \epsilon} + \frac{\partial \ln S}{\partial \epsilon} \right)_{\epsilon_F}, \quad (5)$$

where Λ is the mean free path and S is the area of the Fermi surface. The first term in Eq. (5) is normally positive, because the more energetic electrons are less easily scattered. However, if the s electrons can scatter into a narrow d band with high density of states⁴ then

$$\left(\frac{\partial \ln \Lambda}{\partial \epsilon} \right)_{\epsilon_F} = - \left(\frac{\partial \ln N_d(\epsilon)}{\partial \epsilon} \right)_{\epsilon_F}, \quad (6)$$

where $N_d(\epsilon)$ is the density of states in the d band. This term can be positive or negative.

⁴ J. M. Ziman, *Electrons and Phonons* (Oxford University Press, London, 1960).

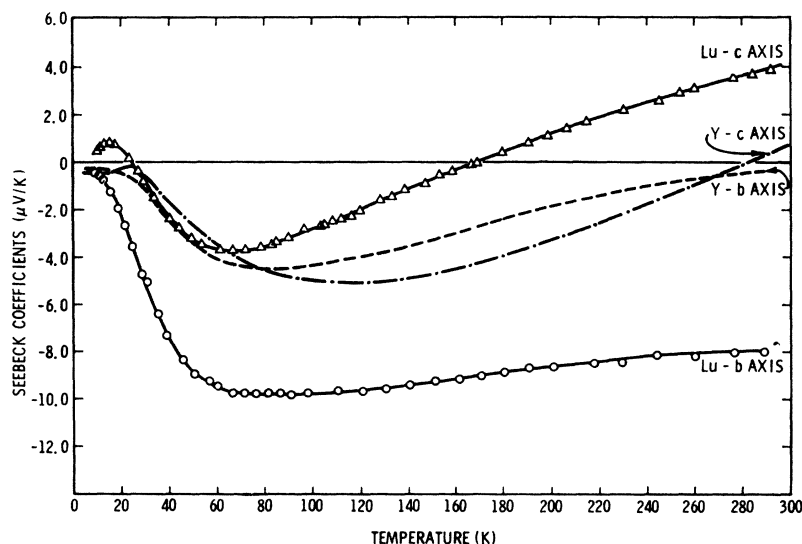


FIG. 1. Seebeck coefficients of lutetium and yttrium single crystals.

Band-structure calculations for the heavy rare earths and yttrium⁵⁻⁸ have been made and indicate that all the Fermi surfaces are very similar and anisotropic. Furthermore, there are many places on the Fermi surface where $\partial \ln S / \partial \epsilon$ can be negative (i.e., holes) and thus give a positive contribution to the Seebeck coefficient. Band-structure calculations⁸ also indicate these materials have a high density of states in the d band around the Fermi energy and thus from Eq. (6) could affect the sign of the Seebeck coefficient. Also thermal conductivity measurements on single crystals of lutetium³ suggest that the phonons carry very little heat along the c axis above 60 K. Thus one would expect above 60 K that the major contribution to the Seebeck coefficient would be from the diffusion component only. To understand the Seebeck coefficients of lutetium in more detail, a theoretical calculation of the diffusion component using the actual Fermi surface will be necessary.

⁵ J. O. Dimmock and A. J. Freeman, Phys. Rev. Letters **13**, 750 (1964).

⁶ A. J. Freeman, J. O. Dimmock, and R. E. Watson, *Quantum Theory of Atoms, Molecules, and the Solid State* (Academic Press Inc., New York, 1966), p. 372.

⁷ T. L. Loucks, Phys. Rev. **144**, 504 (1966).

⁸ S. C. Keeton and T. L. Loucks, Phys. Rev. **146**, 429 (1966).

It is somewhat surprising that there is greater anisotropy in the lutetium Seebeck coefficients than in the yttrium Seebeck coefficients, since band-structure calculations indicate that their respective Fermi surfaces are very similar. Furthermore, the resistivities of lutetium³ and yttrium⁹ are nearly equal in magnitude at room temperature and have nearly identical temperature dependences. Since the Seebeck coefficient is very sensitive to the electronic structure, these observed differences between lutetium and yttrium indicate that there must be subtle differences in their respective Fermi surfaces. Specifically, band-structure calculations indicate that at the Fermi energy for lutetium the density of states is increasing with increasing energy, while for yttrium the density of states is decreasing with increasing energy.^{7,8}

The low-temperature (< 20 K) results must be viewed with some caution because of impurity scattering effects. Although the lutetium used in this study is 99.9% pure there is still sufficient iron to affect the Seebeck coefficient at low temperature.

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⁹ P. M. Hall, S. Legvold, and F. H. Spedding, Phys. Rev. **116**, 1446 (1959).