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Seebeck Effect in Heavy Rare-Earth Single Crystals*

L. R. SILL AND SAM LEGVOLD
Institute for Atomic Research and Department of Physics, Iowa State University, Ames, Iowa

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Seebeck coefficients S have been measured along the three crystallographic directions $\langle 11\bar{2}0\rangle$, $\langle 10\bar{1}0\rangle$, and $\langle 0001\rangle$ for the heavy rare-earth metals Gd, Tb, Dy, Ho, and Er over the temperature range 7 to 300°K. The $\langle 10\bar{1}0\rangle$ and $\langle 0001\rangle$ directions of Y were also measured. In the cases of Gd and Tb, a 1000-Oe field along the $\langle 0001\rangle$ and $\langle 10\bar{1}0\rangle$ directions, respectively, caused no marked change. A significant anisotropy between the $\langle 0001\rangle$ direction and the basal plane was found for all the metals except Y. The magnetic order-disorder temperatures are marked by changes in slope in the S-versus-T curves. A discontinuity in S at the magnetic transition is evident in the case of Dy.

INTRODUCTION

THERMOPOWER measurements provide an excellent method, which is generally superior to resistivity measurements, to study small changes in the state and in the Fermi surface of metals. The Seebeck coefficient S is extremely sensitive both to changes in electronic structure and to the mechanisms which scatter electrons.[1]

The Seebeck coefficients of polycrystalline rare-earth metals have been investigated by Born *et al.*[2] from 7 to 300°K. Interesting magnetic transformations and anisotropies in the heavy rare earths[3] have suggested that the thermoelectric measurements should be extended to single crystals of these metals. Results of such measurements are reported here for Gd, Tb, Dy, Ho, Er, and Y.

The electronic configurations of the heavy rare-earth metals differ from each other only in the number of localized electrons in the $4f$ shell. Their magnetic behavior results principally from these localized $4f$ electrons. For all of the metals measured, there are three conduction electrons which occupy overlapping s (or sp) and d bands at the Fermi energy.

The measurements reported here in most cases provide confirmation of the anomalous behavior noted in heat capacity, resistivity, thermal expansion, and magnetic moment measurements[4] at the magnetic transition temperatures. Because thermoelectric effects depend strongly on the relative concentrations of impurities in the metal,[5,6] particularly at very low temperatures, the reported curves of S versus T must be viewed with some caution. However, it is expected that most of the features reported here will be characteristic of the very pure metals.

EXPERIMENTAL PROCEDURE

The metals used were of the highest purity available at the present time. The single crystals were grown from arc-melted buttons as described by Nigh.[7] It was found that for all of the metals the probability for large grain growth was greatest if the following techniques were used: The arc-melted button was annealed in a temperature gradient of about 25°C/cm starting at a tempera- of 200°C below the melting temperature or transition temperature (whichever was lower), and then the temperature was raised in three successive steps by 50-deg intervals every 8 h. Finally, the button was annealed for about 8 h at the highest temperature in the constant temperature zone before it was slowly cooled down over a 3-h period.

The single crystals thus obtained were checked for orientation by means of a back reflection Laue x-ray camera technique. Three samples were cut by spark erosion from the bulk crystals. These were in the shape of rectangular parallelepipeds such that the long dimension was parallel to the $\langle 0001\rangle$ direction (c axis), $\langle 11\bar{2}0\rangle$ direction (a axis), and $\langle 10\bar{1}0\rangle$ direction (b axis), respectively. The accuracy of this orientation was within 1° of that stated. The samples were about 2×2 mm in cross section and ranged from a minimum of 10 mm to a maximum of 22 mm in length with an average of about 14 mm.

Small pieces of most of the metals were analyzed spectrographically and by vacuum fusion for impurities. In addition, resistivity ratios and residual resistivities (at 4.2°K) were measured by the standard four-probe method on each sample. The most significant results are shown in Table I.

The experimental set-up was basically the same as that described by Born *et al.*[2] However, several modifications were made to accommodate the small sizes of the single crystal samples used. The temperature range from 4.2 to 300°K was covered in three steps instead of two in order to give better low-temperature data. The data were taken in the following order: 80–300°K using liquid nitrogen as the bath, 22–90°K using liquid hydrogen, and finally, 7–30°K using liquid helium.

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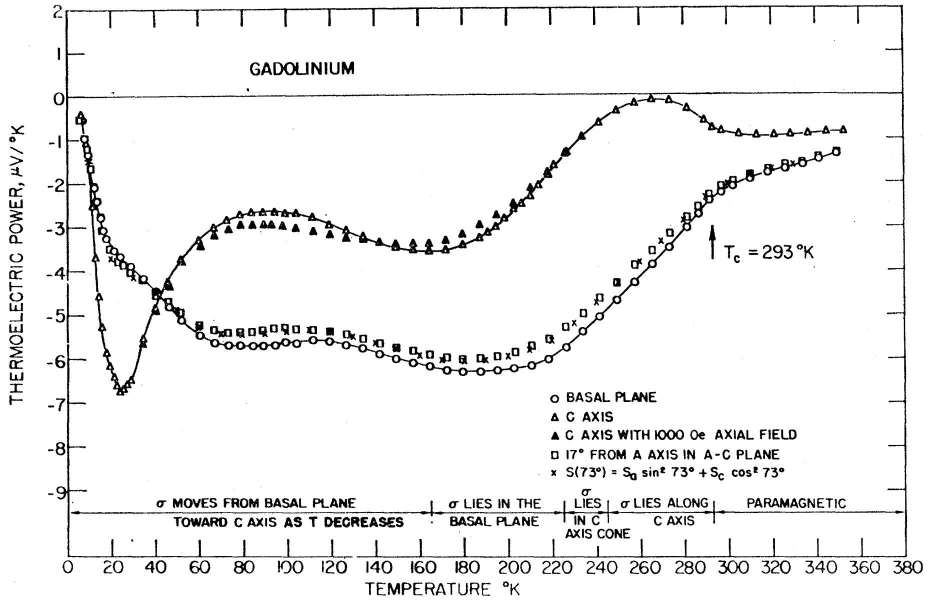

FIG. 1. Seebeck coefficient of gadolinium single crystals versus temperature. σ here is the magnetic moment direction.

One end of the sample was soldered to the heat leak can and the other end was soldered to a small heater used to provide the temperature gradient along the sample. A temperature difference of 2–3°K was maintained except when starting out from liquid-helium temperature. It was possible to solder to the rare-earth metals if they were first tinned by using an ultrasonic soldering gun. Pure indium was used both to tin the sample and to solder it into the sample chamber. Embedded in the solder joint on each end of the sample was a copper-constantan thermocouple. By soldering the sample to the apparatus and by embedding the thermocouples in the solder joints, several possible errors are eliminated: (1) The thermocouple gives the correct temperature at the reference material sample junction, (2) surface layers are eliminated; (3) thermal contact resistance is reduced, and thus the temperature drop at the interface is eliminated. The sample temperature was taken to be the average of those measured at the ends of the sample. The Seebeck voltage V_{SC} was measured between the copper wires of the two thermocouples. The difference voltage between the two thermocouples V_{TC} was measured by means of a Dauphinee potential comparator.[8] The voltages V_{SC} and V_{TC} were measured with a thermofree Rubicon model-2771 microvolt double potentiometer. The absolute Seebeck coefficient of the specimen was determined by standardizing the copper wires with a piece of pure lead whose absolute Seebeck coefficient was well known.[9] By convention, $S_{SC}=S_S-S_C$. This method of measurement is outlined by Heikes and Ure.[10]

TABLE I. Selected sample impurities, residual resistivity and resistivity ratios.

Sample (axis)		Impurities (%) Fe	O_2	Ta	$R_{4.2°}$ (10^{-6} Ω-cm)	$R_{300°}/R_{4.2°}$
Gd	*a*	0.0060	0.2000	0.0350	6.1	22.2
	b	0.0060	0.2000	0.0475	6.3	21.6
	c	0.0060	0.1600	0.1100	2.7	45.0
Tb	*a*	0.0110		0.4000	4.4	24.8
	b	0.0065	0.0850	0.2800	4.1	29.3
	c	0.0065	0.1000	0.5800	4.0	23.6
Dy	*a*				10.0	10.8
	b				7.0	14.9
	c	0.0600	0.0937	0.0300	6.5	11.0
Ho	*a*	0.0100		0.0500	3.0	31.6
	b	0.0075	0.0058	0.0500	2.8	34.5
	c	0.0050		···	4.7	11.7
Er	*a*	0.0100	0.0230	0.0500	5.9	14.1
	b	0.0150	0.0235	0.0500	5.7	14.8
	c	0.0900	0.0280	0.0500	5.2	8.1
Y	*b*	0.0200		0.0400	4.8	14.6
	c				2.1	15.4

The relative accuracy of the measurements is estimated to be within ±0.05 μV/°K for the Seebeck coefficient and within ±0.05°K for the temperature. The systematic errors are more difficult to evaluate, but above 20°K the absolute accuracy of the Seebeck coefficient measurements is estimated to be within ±0.1 μV/°K, and that of the temperature to be within ±0.5°K. Below 29°K the data are somewhat less accurate because of loss of sensitivity of the thermocouples. The error in the residual resistivity is estimated to be $\pm 0.1\times 10^{-6}$ Ω cm and that of the resistivity ratio ±0.4.

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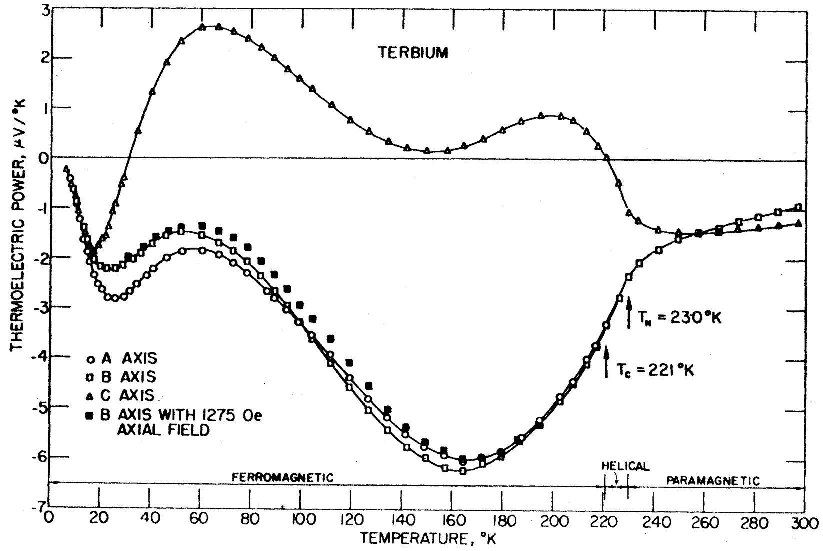

FIG. 2. Seebeck coefficient of terbium single crystals versus temperature.

RESULTS

The observed temperature variations of the Seebeck coefficients of the heavy rare earth metals and of yttrium are shown in Figs. 1–6. In each figure just above the temperature axis the magnetic ordering in the metal is indicated: the moment direction for Gd has been reported by Graham,[11] by Corner *et al.*,[12] and by Will *et al.*[13]; for Tb by Koehler *et al.*,[14] for Dy by Wilkinson *et al.*,[15] for Ho by Koehler *et al.*,[16] and for Er by Cable *et al.*[17] A pictorial summary of the magnetic ordering for the latter four metals is shown in Fig. 7.

Magnetic ordering effects appear to be responsible for the complexity of the data for the heavy rare-earth metals. This conclusion is based on a comparison of the results in Figs. 1–5 with the results for yttrium in Fig. 6. It seems proper to make this comparison because yttrium is a trivalent hcp metal with lattice constants very close to those of Gd and with a polycrystalline Seebeck coefficient very similar to that for Lu.[2]

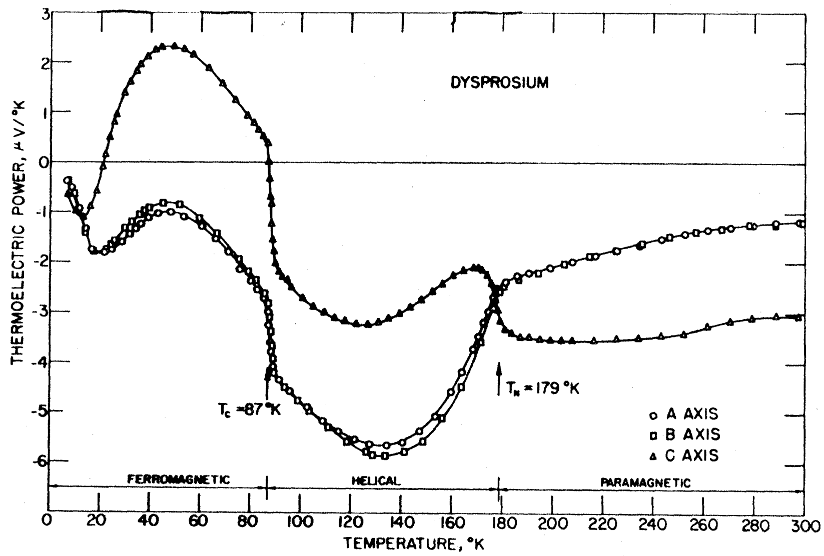

FIG. 3. Seebeck coefficient of dysprosium single crystals versus temperature.

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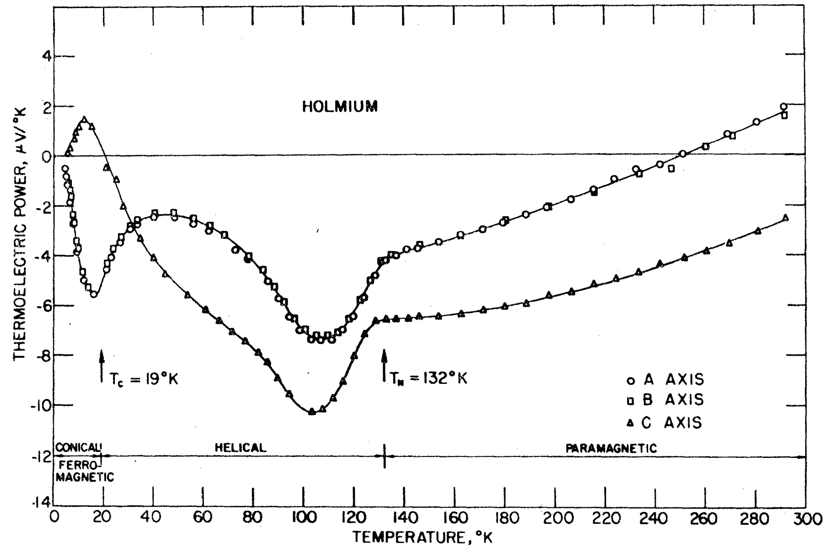

FIG. 4. Seebeck coefficient of holmium single crystals versus temperature.

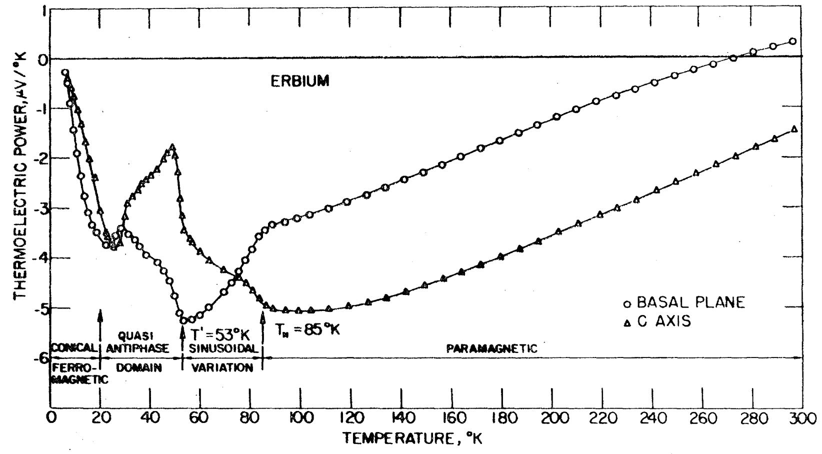

FIG. 5. Seebeck coefficient of erbium single crystals versus temperature.

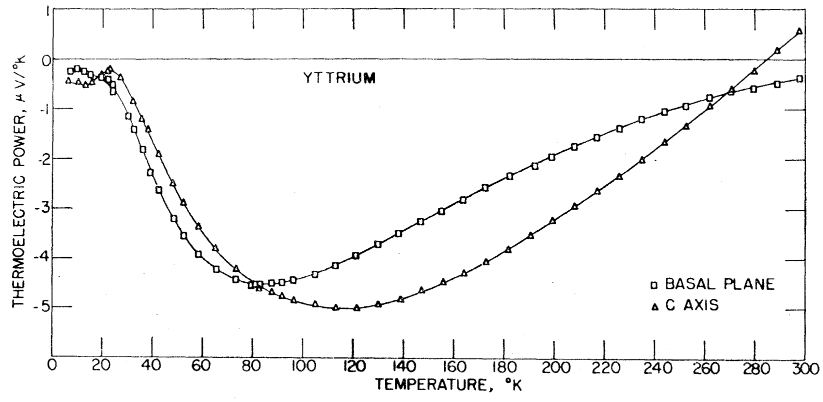

FIG. 6. Seebeck coefficient of yttrium single crystals versus temperature.

For the metals Tb, Dy, Ho, and Er, the *c*-axis curve lies below the basal plane curve in the paramagnetic region. Above the magnetic order-disorder temperature the absolute Seebeck coefficient increases almost linearly in all the metals, except possibly Dy, and appears to become positive at higher temperatures (above 300°K). At low temperatures, the Seebeck coefficient for all of the metals appears to be negative as the temperature approaches 0°K. The magnetic order-disorder temperatures are marked by changes in slope in the S versus

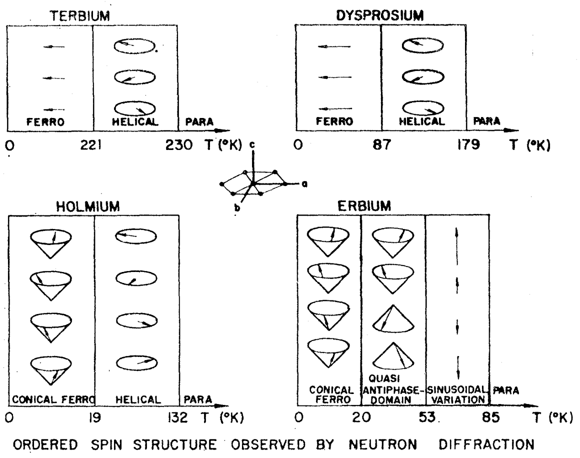

FIG. 7. Ordered spin structure observed by neutron diffraction.

T curves. For all of the heavy rare-earth metals studied except Ho, the c-axis S becomes less negative with decreasing temperatue immediately below the order-disorder temperature; in Ho, the c-axis S becomes more negative. In Tb, Dy, and Ho, the c-axis S becomes positive at the Curie temperature. The magnetic transition from antiferro- to ferromagnetism is obvious only in the case of Dy where a precipitous change is observed. A considerable anisotropy in S occurs in the basal plane only where the magnetization behavior in the basal plane is anisotropic, i.e., only for Dy and Tb.

The effect on S of a 1000-Oe external magnetic field along the c axis of Gd was modest, as can be seen from the results shown in Fig. 1. An observable effect was also found along the b-axis (easy) direction in Tb (see Fig. 2), but there was no observable effect when the field was applied along an a-axis sample. In the case of the Tb b-axis sample, a hysteresis in S of magnetic origin was observed. Only the 1000-Oe external field results were reproducible unless the sample was magnetically annealed between data points.

Among the results for Gd shown in Fig. 1 are data taken on a sample aligned 73° away from the c axis in the a-c plane together with values calculated for such a crystal from the principal axes data. Good agreement is indicated.

DISCUSSION

Although a large amount of work has been done on the Seebeck coefficient in solids,[18,19] the effect of magnetic ordering on the Seebeck effect has not been extensively studied. It has been known for many years, however, that S behaves anomalously near the ferromagnetic Curie point,[20-22] and more recently anomalies have been observed near the Néel temperature.[23,24] The interactions between the conduction electrons and the localized magnetic moments and their effect on the transport properties have been considered by Rezanov[25] and Kasuya.[26] The role of magnon-electron scattering in the Seebeck coefficient of ferromagnetic metals has been studied by Bailyn[27] and more recently by Gurevich and Nedlin.[28] The rare-earth metals can have complex magnetic structures, however, and these can profoundly affect the transport properties through the spin-wave spectra[29] and the modification of the Fermi surface by magnetic Brillouin zone boundaries.[30,31] No detailed theory of these effects has yet appeared.

The Seebeck coefficient of a magnetic material is believed to arise from three main sources: (1) diffusion of electrons, (2) phonon drag, and (3) magnon drag. As a first approximation, these three contributions can be considered independently, so that

$$S=S_e+S_{\mathrm{ph}}+S_{\mathrm{mag}}. \tag{1}$$

At high temperatures, the electron diffusion term predominates and it is the effect of the magnetic ordering on this term which can be observed in the results near the ordering temperatures. It has been suggested by Mackintosh,[30,31] Miwa,[32] and Elliott and Wedgwood,[33] that in the antiferromagnetic state there are extra planes of energy discontinuity introduced into the Brillouin zone structure as a result of the periodic arrangement of magnetic moments along the c axis, which is incommensurate with the ionic lattice. Hence, new energy gaps appear in the electron dispersion curve at the ordering temperatures and increase in magnitude as the temperature is lowered.

The Seebeck coefficient due to electron diffusion is given by the general expression

$$S_{ij}=\frac{\pi^2k^2T}{3e}\left[\frac{\partial\log\sigma_{ij}(\epsilon)}{\partial\epsilon}\right]_{\epsilon=\epsilon_F}, \tag{2}$$

where

$$\sigma_{ij}=\frac{e^2}{4_{\pi}2_{h}}\int_{\epsilon}\tau V_i d\Sigma_j\,, \tag{3}$$

and where k is Boltzmann's constant, T is the absolute temperature, e is the electronic charge, σ_{ij} is the elec-

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trical conductivity, V_t is a component of electron velocity on an energy surface element $d\Sigma_j$, τ is the relaxation time, and the integral is taken over a constant energy surface ϵ. The total relaxation time can be written in terms of the impurity, lattice, and magnetic relaxation times as

$$1/\tau = 1/\tau_i + 1/\tau_e + 1/\tau_m. \tag{4}$$

Mackintosh[34] then takes the simple model of Miwa[35] and Elliott and Wedgwood[33] in which a spherical Fermi surface is cut by a single energy gap of magnitude Δ, allows the magnetic relaxation time to have an energy dependence given by

$$1/\tau_m = \alpha\epsilon^n \tag{5}$$

and neglects the energy dependence of the lattice and impurity relaxation time, and finds

$$S_z = \frac{LeT}{2\epsilon_F}\left(\frac{3}{1-\delta} - \frac{2n\tau}{\tau_s}\right), \tag{6}$$

$$S_x = \frac{LeT}{2\epsilon_F}\left(3 - \frac{2n\tau}{\tau_s}\right), \tag{7}$$

where L is the Lorenz number and δ is approximately Δ/ϵ_F. For the simple model of Kasuya[36] and DeGennes and Friedel[37] n is $\frac{1}{2}$.

We consider first the results for Dy. The rapid decrease of S below the Néel temperature in the basal plane can be attributed to the rapid decrease in the magnetic scattering as the temperature is lowered in this region. The same effect takes place in the c direction but is overcome near T_N by the increase in S due to the superzone energy gaps. The relative signs of these effects indicates that n is negative in this case. A similar explanation can be invoked to explain the anomalies near the Néel temperatures of the other metals, although the effects are not so clear, and in holmium the effect of the energy gaps appears to be small. The small bump in the c-axis results for Gd near T_c suggests the possibility of a more complicated magnetic structure in this region.

At low temperatures the electron diffusion term is expected to become small and the phonon and magnon drag contributions dominate the results. At low temperatures S_e is proportional to T, while S_{ph} is proportional to T^3 up to a maximum between $\theta_D/5$ and $\theta_D/10$ above which it falls as T^{-1}. According to Bailyn[27] S_{mag} should behave similarly to the phonon drag term. The Debye θ_D for the metals studied ranges from 152°K for Gd to 163°K for Er, and one would therefore expect the phonon drag maxima to be between 15 and 30°K. The results for Y, in which there is only a rather small bump at low temperatures, indicate that phonon drag may be rather small in these specimens and that the large bumps in the Seebeck coefficients of the magnetic metals at low temperatures may be due to magnon drag. The magnon contribution might also explain the large Lorenz numbers found for Dy[38] and Gd.[39] In Fig. 8, we have plotted S/T versus T^2 for Gd at low temperatures and find a straight line as expected for either phonon or magnon drag at low temperatures. The other heavy rare-earth metals are magnetically anisotropic so that the lowest lying spin wave state has a nonzero energy as suggested by Niira.[40] In Dy and Tb, these energy gaps in the spin-wave spectra are equivalent to about 20°K, and it is noteworthy that in these metals a rapid change in the Seebeck coefficient occurs at around this temperature. This is probably due to a sudden onset of magnon drag at this temperature, although it is possible that the electron diffusion term is also enhanced by spin-wave scattering of the electrons. Ho and Er have smaller spin-wave energy gaps and low-temperature magnetic transitions, and these effects cannot be so clearly observed.

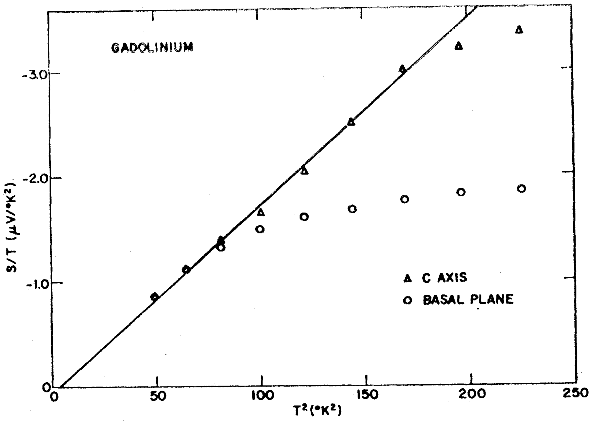

FIG. 8. Phonon-magnon drag in gadolinium single crystals.

The only discontinuity observed in these results occurs at the transition from a helix to a classical ferromagnet in Dy. Since this does not occur in the other metals, we believe that it must be due to the transition from a hexagonal to an orthorhombic structure at T_c.[41] Of the other metals measured in this study Gd, Ho, and Er show no orthorhombic distortion in the ferromagnetic state.[42,43] In Tb, the orthorhombic distortion at T_c is very small because the sublattice magnetization is small.[39] This distortion increases as the sublattice magnetization increases with decreasing temperature,

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and consequently there is no sudden change and no discontinuity in S. The appearance of superzone energy gaps and the change in the spin wave spectrum at the transition could also contribute to the discontinuity in Dy, but we believe that the primary effect is due to the crystal structure change; the observed sizeable lattice parameter changes[42,43] could have a significant effect on the Seebeck coefficients.

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Doppler-Shifted Cyclotron Resonance with Helicon Waves*

M. T. TAYLOR†
Laboratory of Atomic and Solid State Physics, Physics Department, Cornell University, Ithaca, New York

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The properties of helicon waves have been studied under the condition that the electron mean free path is larger than the helicon wavelength; this is often called the nonlocal limit. The free-electron theory shows that under this condition there is a threshold magnetic field. Above this threshold, undamped helicon waves can propagate in the metal. Below it, the wave is heavily damped by cylotron resonance of the electrons. This threshold field has been called the Kjeldaas absorption edge or Doppler-shifted cyclotron resonance. At fields just above the edge, the dispersion relation is modified from the simple form appropriate for the local limit. At the edge there is a singularity in the surface impedance of the metal sample. The theory of these effects has been tested by measurements on polycrystalline sodium and potassium, using two experimental techniques. First, the dispersion relation was studied using helicon waves propagating through a metal slab. Second, the surface impedance was studied at radio frequencies. Excellent agreement between theory and experiment was obtained. The values for the radius of the Fermi surface were $(0.92\pm0.01)\times10^{+8}$ cm^{-1} for sodium and $(0.74\pm0.01)\times10^{+8}$ cm^{-1} for potassium. These are to be compared with the free-electron values of $0.923\times10^{+8}$ cm^{-1} and $0.746\times10^{+8}$ cm^{-1}, respectively. The sharpness of the edge as a function of the electron mean free path was also studied. It was found that the fractional width was essentially inversely proportional to $\omega_c\tau$. The edge has also been studied with polycrystalline indium.

I. INTRODUCTION

HELICON waves are transverse electromagnetic waves which, in the presence of an applied magnetic field, can propagate in a metal. For a free-electron metal, with n electrons per unit volume, in a field B, the helicon wave dispersion relation is[1]

$$q^2=\mu_0 ne\omega/B(mks\text{ units})\,, \qquad (1)$$

where q is the wave vector for a wave of frequency ω and μ_0 is the permittivity of free space.

Helicon waves have been extensively studied, especially in the region where the wavelength is greater than the electron mean free path; both standing wave and transmission experiments have been reported.[2] These experiments can be used to determine such properties of the metal as the Hall coefficient and the magnetoresistivity.

This paper is concerned with the study of helicon waves when the electron mean free path ℓ becomes much larger than the wavelength, that is when $q\ell\gg1$. Under this condition a study of helicon waves can yield information concerning the geometry of the Fermi surface. In contrast to the case when $q\ell\ll1$, it is no longer sufficient to specify the electron's behavior in terms of an average drift velocity. For $q\ell\gg1$ it becomes necessary to consider the details of the electron's orbit and its interaction with the wave. Such considerations lead to the prediction of cyclotron resonance at frequencies very much smaller than the cyclotron frequency. This occurs because an electron at the Fermi surface travels through the slow helicon wave and experiences, as a consequence of the Doppler effect, an electric field having an apparent frequency very much larger than the actual wave frequency. If the apparent frequency equals the electron's cyclotron frequency, Doppler-shifted cyclotron resonance occurs; the electron then absorbs energy from the wave.

The effect was predicted by Kjeldaas[3] for transverse ultrasonic waves propagating in a metal parallel to a

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† Present address: Central Electricity Research Laboratories, Cleeve Road, Leatherhead, Surrey, England.

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