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Righi–Leduc Effect in Indium*

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Measurements of the magnetic field and temperature dependence of the Righi–Leduc coefficient have been made in pure oriented single-crystal specimens of indium in the temperature range 3.5–8 K. The coefficient is observed to change from negative at low fields to positive at sufficiently large magnetic fields. The value of the crossing field, which is on the order of a few kG, increases with temperature. Analysis of the results in terms of a two-band model for the electronic carriers yields an estimate of about 1.2 for the ratio of electron-to-hole relaxation times.

I. INTRODUCTION

The Righi–Leduc (RL) effect is the thermal analog of the Hall effect. For a metal carrying an electronic heat current U_x in the x direction, in the presence of an applied magnetic field H in the z direction, the RL coefficient R_L is defined as

$$R_L = \nabla T_y / H U_x ,$$

where ∇T_y is the temperature gradient transverse to both the heat current and the magnetic field. In the high-field limit, the Hall and RL effects in metals having a closed Fermi surface are found both theoretically[1] and experimentally[2,3] to be determined solely by the carrier densities and charges. For such a metal with two bands of carriers of opposite sign and densities n_1 and n_2, these coefficients are given by

$$R_H = 1/(n_1 - n_2)ec , \quad R_L = 1/L_0 T(n_1 - n_2)ec ,$$

where e is the magnitude of the electronic charge, T is the absolute temperature, $L_0 = \pi^2 k^2/3e^2 = (2.45 \times 10^{-8})\ \mathrm{W\Omega/K^2}$ is the free-electron Lorentz number, and k is the Boltzmann constant. The Hall coefficient R_H and the RL coefficient are related to each other through the Wiedemann–Franz law, even in the presence of inelastic scattering.

At low fields, however, the detailed topology of the Fermi surface and the various mechanisms by which the charge carriers are scattered become important in determining these effects. Thus, when the Fermi surface of a particular metal is known accurately, measurements of the Hall and RL effects can yield information about the carrier relaxation times. Under conditions where the Wiedemann–Franz law is not obeyed, that is, when the electrical and thermal relaxation times are not the same, measurements of the Hall and RL coefficients each give different information about scattering effects. Estimates of the electron-hole relaxation-time ratio in single-crystal specimens of indium have been made from Hall-effect data.[4,5] In this paper we present the results of RL-effect measurements in oriented single-crystal specimens of indium, over the temperature range 3.5–8 K and in magnetic fields up to 15 kG. There are important differences between the electrical and thermal results; these are discussed after the data are presented.

II. THEORY

The Fermi surface of indium consists of a hole-like surface in the second zone and an electronlike one in the third zone. The simplest model that might be expected to yield qualitatively correct predictions of the magnetic field and temperature dependence of the RL effect in this material is one in which the metal is considered to have two overlapping parabolic bands of carriers in which interband transitions are neglected. It is assumed that for each band scattering processes can be described by a single isotropic relaxation time; this

relaxation time can be different for each band, but it is assumed constant within a band. For this case, the RL coefficient can be written

$$R_L = \frac{\sum_i [R_{Li}\kappa_i^2/(1+\omega_i^2\tau_i^2)]}{\{\sum_i [\kappa_i/(1+\omega_i^2\tau_i^2)]\}^2 + \{\sum_i [\kappa_i\omega_i\tau_i/(1+\omega_i^2\tau_i^2)]\}^2}, \quad (1)$$

where $\omega_i = e_i H/m_i c$ is the cyclotron frequency for a carrier of charge e_i and mass m_i. Here R_{Li} and κ_i are, respectively, the RL coefficient and the thermal conductivity of the ith band.[6] While this crude model of the Fermi surface precludes any reliable quantitative predictions, some important qualitative features are evident. In particular, if the R_{Li} have opposite signs, there exists the possibility of a reversal in sign of R_L as the magnetic field (and hence $\omega_i\tau_i$) is increased from zero. Reversal arises from the fact that the small-mass carriers reach the high-field limit ($\omega_i\tau_i \gg 1$) before the heavier carriers.

With the previous assumptions, the RL coefficient and thermal conductivity for each band can be expressed in terms of the corresponding Hall coefficient and electrical conductivities by the usual relationships:

$$R_{Li} = 1/LTn_i e_i c, \quad \kappa_i = LTn_i e_i^2 \tau_i/m_i .$$

In the case where electrical and thermal relaxation times are the same, the parameter L assumes the value of the free-electron Lorentz number L_0. We consider the case of carriers of opposite sign [electrons (e) and holes (h)]. Denoting the value of the magnetic field at which $R_L = 0$ (the crossing field) by H_0, we find upon substituting the above expressions into Eq. (1) that H_0 is given by

$$H_0 = \frac{m_h c}{e}\left(\frac{n_e}{n}\right)^{1/2}\tau_h^{-1}\left(1 - A\frac{\tau_h^2}{\tau_e^2}\right)^{1/2}, \quad (2)$$

where $A = n_h m_e^2/n_e m_h^2$ and $n \equiv n_h - n_e$ is the net carrier density (assumed positive). This model then predicts that a magnetic-field-dependent reversal in sign of the RL coefficient will occur if the relation

$$\frac{n_h}{n_e}\left(\frac{m_e\tau_h}{m_h\tau_e}\right)^2 < 1 \quad (3)$$

is satisfied.

In the case of indium, the holelike carriers of the second zone have a density of about one hole per atom and a cyclotron effective mass of about the free-electron mass. The third zone consists of electronlike carriers, with a density of a few percent of an electron per atom and an effective mass on the order of 0.1 times the free-electron mass. For roughly equal hole and electron relaxation times, a sign reversal in indium is expected from Eq. (3). Equation (2) predicts a crossing field on the order of a few kG for relaxation times of roughly 10^{-12} sec; this is consistent with what is observed in indium.

In order to obtain better than an order-of-magnitude estimate, the actual Fermi-surface topology must be taken into account. The model that will be used in evaluating our experimental results is based on one suggested by Ashcroft,[4] which was used for the interpretation of Hall-effect data.[5] The model assumes two noninteracting bands of carriers. Each band has a distinct relaxation time, but within each band the relaxation time remains constant. The nonsphericity of the Fermi surface is taken into account by noting that, while the carrier velocities are not constant over a cyclotron orbit, they are nevertheless periodic. Thus, one can make a Fourier expansion of carrier velocities of the form

$$v_i = \sum_n e^{jn\theta} v_{in},$$

where θ is a phase variable proportional to time and $j = (-1)^{1/2}$. For a magnetic field in the z direction and normal to planes of reflection symmetry in the crystal, the condition necessary for the RL effect to vanish is

$$\tau_h^2 \int \frac{dk_z}{k_F} \sum_n \frac{n v^h_{x,n} v^h_{y,n}}{1+n^2\omega_h^2\tau_h^2} = \tau_e^2 \int \frac{dk_z}{k_F} \sum_n \frac{n v^e_{x,n} v^e_{y,n}}{1+n^2\omega_e^2\tau_e^2} . \quad (4)$$

The Fourier velocity components v_{in} can be obtained from a combination of band-structure calculations and Fermi-surface measurements (for example, de Haas–van Alphen-effect measurements). If the crossing field occurs before either carrier is in the high-field limit, then Eq. (4) can be used to determine τ_e/τ_h, the relative relaxation times of the two bands.

Taking into account the known Fermi-surface topology of indium, Ashcroft predicts that a Hall field reversal will occur when the magnetic field is along the [100] or [010] crystal axis, but not when along the [001] axis. In contrast, aluminum, which has fcc rather than face-centered-tetragonal (fct) crystal structure, is expected to show a Hall field reversal for all field directions.

These predictions are supported to some extent by experimental results, in that a sign reversal of the Hall effect has been observed in bulk polycrystalline aluminum,[7–9] but not in bulk polycrystalline indium.[5,9–11] However, indium single crystals of certain orientations[5] and indium specimens in which the carrier mean free path is size-effect limited[10] do exhibit a sign reversal.

In the case of the RL effect, the situation is somewhat different. A sign reversal has been observed not only in single and polycrystalline specimens of aluminum, but in bulk polycrystalline in-

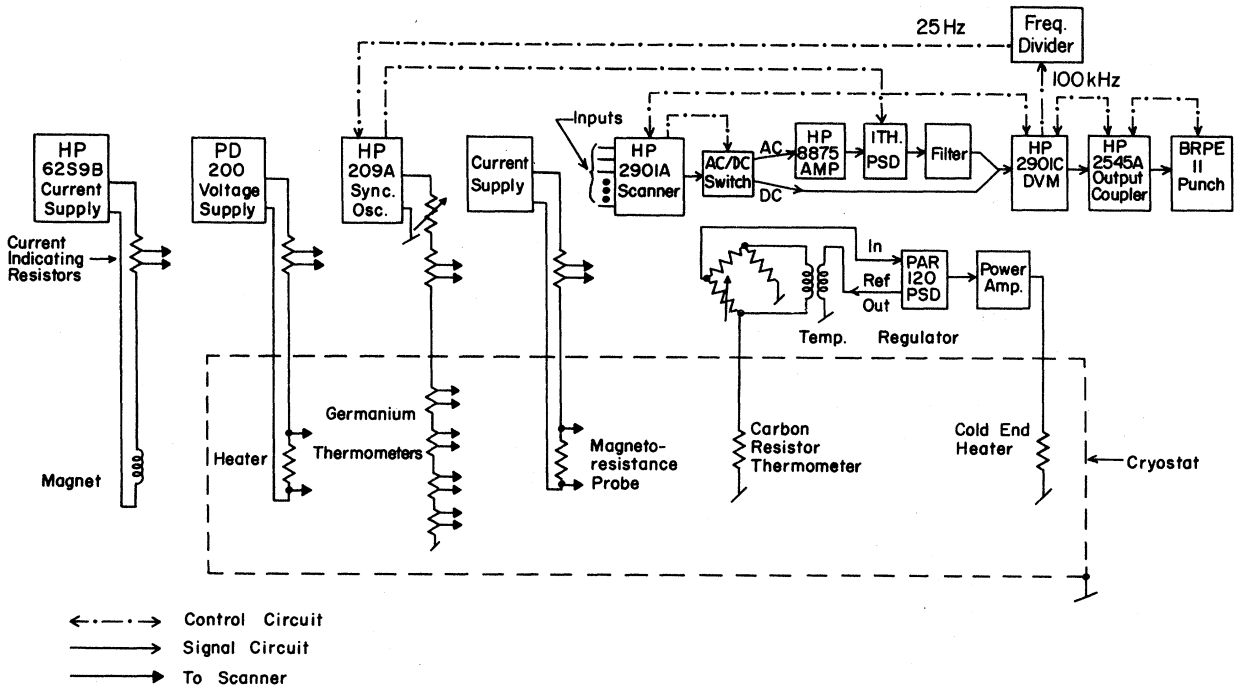

FIG. 1. Block diagram of the instrumentation; HP refers to Hewlett Packard, PD to Power Design, ITH to Ithaco, and PAR to Princeton Applied Research.

dium[2,3] as well. At the temperatures involved, the Wiedemann-Franz law does not hold for indium. Thus it appears that there is a rather delicate balance between the contributions to the Hall and RL resistivities from the two bands—one that, in the thermal case, appears to be tipped in favor of the third-zone electrons when inelastic (phonon) scattering dominates.

III. EXPERIMENTAL TECHNIQUES

A. Instrumentation

A block diagram of the instrumentation used in the experiment is shown in Fig. 1. Doped-germanium resistance thermometers were used to measure the small temperature differences induced in the indium specimens. Carbon resistance thermometers, with their lower magnetoresistance, would have reduced some of the problems associated with using a thermometer in large magnetic fields. However, the superior temperature reproducibility of the germanium resistance elements, and the availability of a data-acquisition system for computerized data analysis, prompted the choice of the germanium thermometers. Their resistance was measured using ac techniques in order to eliminate spurious effects caused by thermal voltages. The resistors were calibrated *in situ* against magnetic field for each temperature of interest. This calibration was repeated every time the thermometers were cycled to room temperature and back. Magnetic fields were generated with an NMR-calibrated superconducting solenoid,[12] and "forward" and "reverse" fields were reproduced accurately with the aid of a bismuth magnetoresistance probe. The data analysis program made corrections for the magnetoresistance of the germanium thermometers. In this way temperature differences of 100 μK could be measured reliably in fields up to 15 kG.

The indium specimen was placed inside an evacuated cryostat with one end linked to the outside liquid-helium bath through a thermal resistance. By means of a feedback system and a heater attached to this colder end, the specimen could be kept at any desired temperature between 2 and 14 K. A heater at the other end of the specimen maintained the thermal current through it. Two pairs of germanium thermometers were held in a precise vertical orientation and pressed lightly against the indium specimen by a small spring pressure. These thermometers measured the thermal gradients along, and transverse to, the direction of heat flow. These gradients were typically no more than 10 mK/cm over the distance of 1–2 cm separating the thermometers.

B. Sample Preparation

The starting material for the specimens used in this experiment was 69-grade indium obtained from Cominco, Inc. The samples were prepared

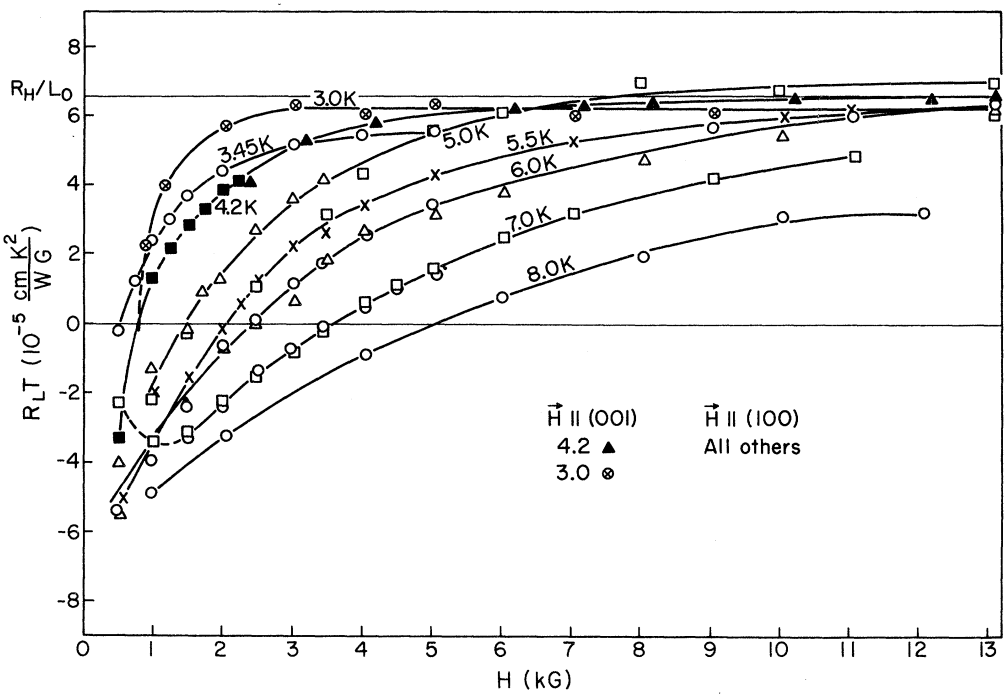

FIG. 2. RL coefficient times absolute temperature as a function of magnetic field between 3 and 8 K.

by melting an ingot into a chlorine-baked carbon crucible under a vacuum of about 10^{-6} Torr. Slow cooling of the crucible under a slight temperature gradient produced a single crystal that was usually oriented with a [001] or [100] axis near the cylinder axis. An oriented plate ($40\times8.5\times1.5$ mm) was cut from the single crystal using an acid saw. Most of the measurements were made on a specimen having a [100] direction perpendicular to the plane of the specimen and either a [100] or [001] direction along the specimen length (that is, those directions where a sign reversal is expected). Some preliminary measurements were also made on a specimen having the [001] direction perpendicular to the plane and a [100] direction along the length. Orientation was accurate to better than 2 deg. The room temperature to 4.2 K resistance ratio (RRR) for all specimens was about 22 000. From this ratio and the size-effect data of Wyder,[13] the electrical mean free path at 4.2 K is estimated to be about 0.3 mm, while that for thermal carriers is about three times smaller. Thus, no size effects are expected in our specimens.

IV. RESULTS AND DISCUSSION

Figure 2 shows the results of RL-effect measurements on indium for various temperatures. The first-order temperature dependence contained implicitly in Eq. (1) has been removed by multiplying all measured temperature gradients by the absolute temperature T. Previous measurements on polycrystalline indium specimens showed that between 2.5 and 8 K the RL effect follows closely a universal curve when the magnetic field is scaled by the inverse of the zero-field electrical resistivity $\rho(T, 0)$.[3] This is somewhat analogous to a Kohler plot of the electrical magnetoresistance; as is customary, the magnetic field variable is taken to be $H\rho(\Theta_D)/\rho(T, 0)$, where $\rho(\Theta_D)$ is the electrical resistivity at the Debye temperature. Much poorer agreement is obtained if the zero-field thermal resistivity is used as the magnetic field scaling factor.[14]

A Kohler plot of the RL effect in single-crystal specimens of indium is given in Fig. 3(a). For comparison, Fig. 3(b) shows the results of earlier measurements[3] on polycrystalline indium specimens having a RRR of 18 000. The polycrystalline data fall reasonably well on a single curve, unlike the single-crystal data, which do not scale with the zero-field electrical resistivity.

To explain this absence of scaling we note that Kohler's rule is predicated upon the existence of a single isotropic relaxation time. Two reasons might be advanced as to why this condition should be satisfied in polycrystalline but not in single-crystal specimens. First, a significant source of elastic scattering present in the polycrystalline case (that due to grain boundaries) is eliminated in the single-crystal specimens. This effect is not likely to be very important, however, because

above 4 K phonon scattering dominates in all single and polycrystalline specimens that were measured. Second, even if the mean free path of the carriers in indium is not isotropic (the degree of anisotropy varying with the amount of phonon scattering, say), one might still expect Kohler's rule to hold in a polycrystalline specimen, in which all crystal directions would contribute equally. This would not occur, of course, for a single crystal.

In order to analyze the behavior of the RL effect in terms of the relaxation times of the carriers in each band, we express the RL coefficient in terms of the electrical resistivity tensor element ρ_{xy} as

$$R_L T = \rho_{xy}/LH.$$

Once again, L is a parameter that assumes the value L_0 under conditions when the Wiedemann–

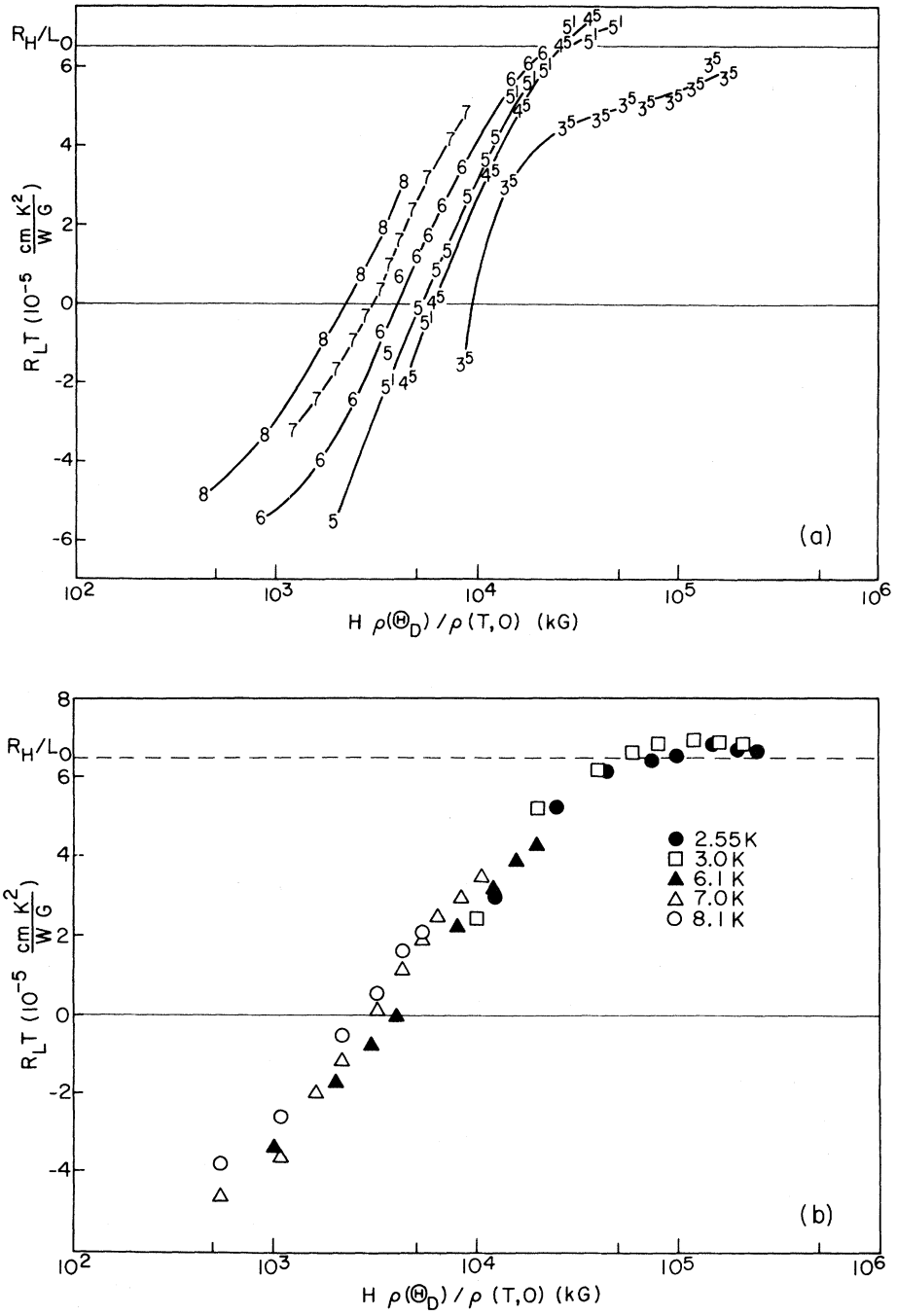

FIG. 3. Reduced-field plot of the RL effect in single-crystal indium. In (a), the numbers refer to the temperature data in Fig. 2 with $\vec{H} \parallel [100]$.

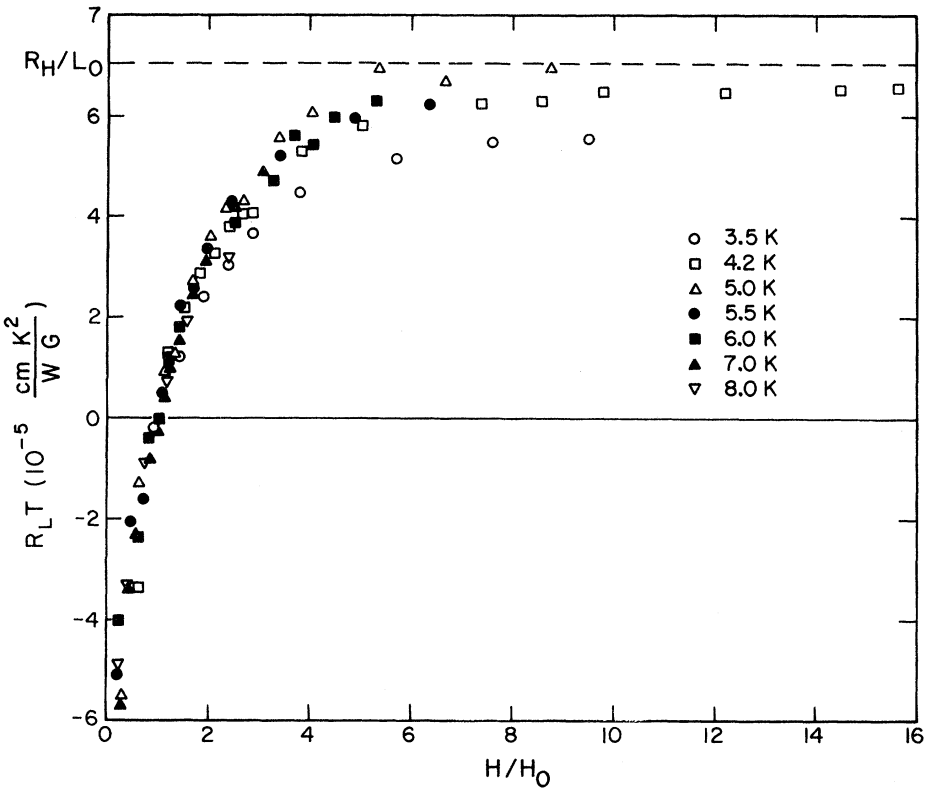

FIG. 4. Magnetic field dependence of the RL effect between 3 and 8 K in single-crystal indium normalized to the crossing field $H_0(T)$.

Franz law holds.

Defining a reduced resistivity $\vec{\vec{\rho}}'$ by

$$(m/ne^2\tau_h)\vec{\vec{\rho}}' = \vec{\vec{\rho}},$$

we have

$$R_L T = \frac{1}{Lnec}\left(\frac{\rho'_{xy}}{\omega_0\tau_h}\right);$$

that is,

$$R_L T = (1/Lnec)F(\omega_0\tau_h,\ \tau_e/\tau_h),$$

where $\omega_0 = eH/mc$ is the free-electron cyclotron frequency. $H_0(T)$, the crossing field, depends on temperature only through the implicit temperature dependence of $\omega_0\tau_h$ and τ_e/τ_h. If we assume that L and τ_e/τ_h are both independent of temperature, then a plot of $R_L T$ vs $H/H_0(T)$ should give a universal curve. Such a plot is shown in Fig. 4. Using a simplified model of the Fermi surface of indium and the method of Ashcroft[4] discussed previously, Hammerberg[15] has calculated from this graph that the ratio τ_e/τ_h is about 1.2. This is a factor of 2 larger than the value of 0.6 calculated by Ashcroft[4] from Hall-effect experiments in indium.[5]

The significance of this difference may be seen by noting that at low temperatures scattering of a carrier through any angle θ is effective in reducing the thermal conductivity. This is not true in the electrical case, where the effectiveness of a scattering event in reducing electrical conductivity is weighted against scattering of the carrier through small angles by a factor like $1-\cos\theta$. Thus, the decrease in τ_e/τ_h in going from the thermal to the electrical case suggests that in indium the electrical conductivity of the third-zone carriers is considerably more reduced by small-angle scattering than is that of the second-zone carriers.

The temperature dependence of the RL-effect crossing field is shown in Fig. 5. Above 4 K, $H_0(T)$ varies very nearly quadratically with temperature. This behavior differs markedly from that observed for the Hall effect, where the magnitude of the crossing field was found to decrease with temperature from a maximum value of about 340 G at 1.4 K. This difference in both magnitude and temperature dependence can be explained by examining Eq. (2); the only factors that can differ between electrical and thermal measurements are τ_e and τ_h. From band-structure calculations[16] and Hall-effect measurements[5] in indium, the term $A\tau_h^2/\tau_e^2$ in Eq. (2) is a very nearly unity. ($A\tau_h^2/\tau_e^2$ must be less than unity in order for a sign reversal to occur.) This makes $H_0(T)$ quite sensitive to small changes in the ratio τ_h/τ_e. There is no rea-

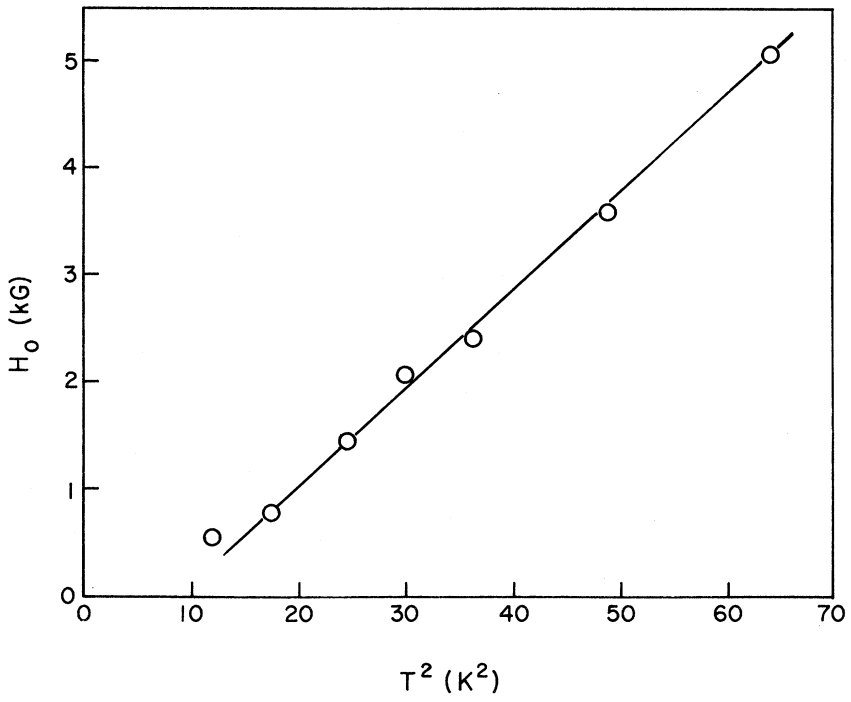

FIG. 5. Temperature dependence of the crossing field $H_0(T)$.

son for τ_h and τ_e to be affected in precisely the same manner as the temperature (and hence phonon scattering) is increased. Even if τ_h/τ_e has a very modest increase with temperature, the $A\tau_h^2/\tau_e^2$ term can dominate the temperature dependence of H_0. This is apparently what happens in the Hall-effect case, where H_0 decreases with increasing temperature; the $1/\tau_h$ prefactor in Eq. (2) plays a relatively minor role in determining the temperature dependence.

From the RL-effect data presented here, it was determined that $(\tau_h/\tau_e)^2 \simeq 0.7$ (compared to about 3 in the Hall-effect case). Hence the $A\tau_h^2/\tau_e^2$ term in Eq. (2) is much smaller for the RL effect; it contributes little to the temperature dependence, which is dominated by the $1/\tau_h$ factor. Since $H_0(T)$ varies quadratically with temperature, $1/\tau_h$ (for thermal scattering) must also vary as T^2. It is difficult to explain a quadratic temperature dependence of any significant magnitude for $1/\tau_h$. Normal electron-phonon–scattering processes result in a cubic temperature dependence. Electron-electron scattering is an obvious candidate; however, estimates of the electron-electron–scattering rate in indium[16,17] give values orders of magnitude too small to explain the observed magnitude of H_0.

The different role of $A\tau_h^2/\tau_e^2$ is also reflected in the magnitude of H_0; when it is near unity H_0 will be small (as in the Hall-effect case). Otherwise, H_0 can become very large because τ_h decreases with increasing temperature.

V. CONCLUSIONS

The RL effect has been measured in single-crystal specimens of pure indium. A Kohler plot of the RL effect in polycrystalline specimens gives a universal curve, whereas an identical plot for single crystals does not. This is attributed to the average scattering being effectively isotropic in the polycrystalline case and anisotropic in the single-crystal case. At high fields and low temperatures where the high-field limit can be attained, the RL coefficient corresponds to the expected value of about 1 hole per atom. At low fields, the RL coefficient depends upon scattering processes as well as the Fermi-surface topology. Above about 3.5 K the condition described by Eq. (3) (or an analogous expression derived by Ashcroft[4]) can be satisfied, and a sign reversal in R_L is observed. From calculations of Hammerberg,[15] the relaxation time ratio τ_e/τ_h is deduced to be about 1.2, a factor of 2 larger than obtained from analogous Hall-effect measurements. When interpreted on the basis of two noninteracting bands of carriers, the quadratic temperature dependence of H_0 is difficult to understand, and requires further clarification.

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Theory of Spin Waves in Nonferromagnetic Metals

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A simple quantum-mechanical theory of spin waves in nonferromagnetic metals has been developed. This theory consists of a description of the spin-wave excitations superimposed on the paramagnetic Hartree–Fock ground state of an interacting electron gas subjected to a constant uniform magnetic field. Use has been made of a gauge-independent density-matrix formalism. It is shown that to first order in the magnetic field the transport equation for the transverse spin magnetization has the same form as the corresponding phenomenological Fermi-liquid equation proposed by Silin and by Platzman and Wolff. It is also shown that for large fields, such that the cyclotron radius is smaller than the interelectronic distance, the term in the transport equation related to the cyclotron motion of the electrons is not affected by the exchange interactions. The theory presented here lends additional credence to the validity of Platzman and Wolff's phenomenological analysis of the experimental spin-wave spectra in the alkali metals.

I. INTRODUCTION

In 1958 Silin predicted the existence of spin-wave excitations in nonferromagnetic metals.[1] These excitations were subsequently observed by Schultz and Dunifer as a series of spin-wave sidebands in conduction-electron spin-resonance experiments performed on sodium and potassium.[2] Platzman and Wolff interpreted the data successfully by means of a phenomenological theory of transport for the transverse spin magnetization in a Landau Fermi liquid.[3] It was shown that this type of experiment provides a means to measure the important Fermi-liquid parameters B_0, $B_1, \ldots$, i.e., those parameters which determine the spin-dependent part $\zeta(\vec{k},\vec{k}')$ of the Fermi-liquid interaction function. Since it is the transport equation for the transverse magnetization itself that will be the subject of this paper, we will reproduce here Platzman and Wolff's transport equation for ease of discussion:

$$\frac{\partial \mu}{\partial t} + \left(\vec{v}\cdot\vec{\nabla} - \frac{e}{\hbar c}(\vec{v}\times\vec{H}_0)\cdot\vec{\nabla}_k - i\Omega_0\right)(\mu + \delta\epsilon_2) = \tfrac{1}{2}\gamma_0^2(\vec{v}\cdot\vec{\nabla} - i\Omega_0)h_+ \,. \quad (1)$$

Here, $\vec{v}$ is the quasiparticle velocity, $\vec{H}_0$ is the applied constant magnetic field (taken in the z direction),

$$\gamma_0 \equiv ge\hbar/2mc, \qquad \Omega_0 = \gamma_0 H_0/(1+B_0)\,,$$

h_+ is the rf magnetic field $h_x + ih_y$, and $\delta\epsilon_2$ is the change in the quasiparticle energy resulting from