Observation of the Saturation of the Longitudinal Thermal Magnetoresistivity of Potassium

R. Fletcher
Physics Department, Queen's University, Kingston, Ontario K7L EN6, Canada
(Received 25 January 1980)

The longitudinal electrical and thermal magnetoresistivities (ρ_{zz} and γ_{zz}) of potassium have been simultaneously measured at fields up to 7.5 T and at temperatures of 2–4 K. Although ρ_{zz} is linear in field, γ_{zz} saturates; this behavior is in agreement with recent predictions of models based on the presence of voids.

PACS numbers: 72.15.He, 72.15.Gd

At high magnetic fields, the electrical resistivity of uncompensated metals (the most prominent being K,[1] Al,[2] and In[3]) increases linearly with magnetic field rather than saturating as theory predicts[4]; this is found to be true for the current either parallel or perpendicular to the field direction B_z (i.e., for either ρ_{zz} or ρ_{xx}). A variety of mechanisms have been proposed to explain the phenomenon, but they have often been presented without a clear appreciation of the fact that any theory must be able to explain all the available results on both compensated and uncompensated metals, and it is known that the latter obey high-field semiclassical predictions very accurately. When viewed from this standpoint,[5] many of the theories are no longer credible.

However, a class of theories that has withstood this test is based on the assumption that inhomogeneities or imperfections in the samples are responsible for the linear behavior.[6-8] As a simplification, these theories usually focus on the effects of voids, and in this case one certainly predicts ρ_{xx} and ρ_{zz} to be linear in field B for uncompensated metals. On the other hand, Stinson[9] has shown that voids would have a negligible effect on ρ_{xx} for compensated metals, a result consistent with their known behavior. Recently the calculations have been extended to the case of the thermal resistivities γ_{xx} and γ_{zz}, independently by Esposito, Newrock, and Loeffler[10] and by Stinson,[9] with the key prediction that the linear component should saturate at a sufficiently high field. The saturation is caused by the lattice conductivity λ_g which prevents the diagonal components, λ_{xx} and λ_{yy}, of the thermal conductivity tensor from decreasing indefinitely as the field increases. For free electrons at high fields, these components take the form $\lambda_0(\omega\tau)^{-2}+\lambda_g$, where λ_0 is the zero-field thermal conductivity of the electrons and $\omega\tau$ is the product of their cyclotron frequency and relaxation time. Thus one expects saturation to occur at a field B_s satisfying the relation $\omega\tau \sim (\lambda_0/\lambda_g)^{1/2}$, which corresponds to about 1 T for K, and 10 T for Al and In. For limiting low values of the fractional volume of voids f, the theories generally predict results of the form $\Delta\rho/\rho_0 = \alpha f\omega\tau$, where $\Delta\rho$ is the change in ρ_{xx} or ρ_{zz}, ρ_0 being the zero-field value, and α a constant of order unity; thus we expect a saturation value of $(\Delta\gamma/\gamma_0)_s \simeq \alpha f(\lambda_0/\lambda_g)^{1/2}$. This prediction of saturation is of great importance because no other theoretical model distinguishes between the electrical and thermal resistivities in this way. The predictions are difficult to confirm experimentally for γ_{xx} because λ_g causes this coefficient to increase approximately as B^2 and this behavior overwhelms the linear term; recent results are consistent with the disappearance of the linear term but are not conclusive.[11] There is no similar B^2 contribution to γ_{zz} but in this geometry the experimental problems are more severe and, in particular, one must take great care to eliminate current jetting.[6,12,13] Initial data by Tausch, Newrock, and Mitchel[14] indicate no saturation of γ_{zz} for K up to about 8 T, but they describe their results as preliminary (see comments later). The data that I shall present have been obtained after an extensive program of tests designed to eliminate spurious effects and, although ρ_{zz} remains linear in B, all of the samples exhibit saturation effects in γ_{zz}.

All samples were cast in the shape of rectangular prisms about 60 mm long with cross sections in the range 3.5–16 mm^2. The last 10 mm at the ends of each sample had a larger cross section (about 5×5 mm^2) to facilitate mounting of the end contacts described below, and two limbs (about $1.5\times1.5\times15$ mm^3) were cast as an integral part of the samples to enable the potential and temperature differences to be measured over the central 19-mm section. Because we require both ρ_{zz} and γ_{zz} for comparison purposes, the only possible method of producing uniform current contacts for both the electrical and heat currents is that of intimate metallic contact over the end faces. After attempting various methods, includ-

ing casting the sample into the end contacts, I developed the following technique which is a modification of that used by Lass.[6] The current source and sink are made of copper, to which 0.75-mm-thick brass sheets have been silver soldered (with sheet solder to eliminate bubbles). The brass faces are grooved in two perpendicular directions to produce an array of pyramids, each about 0.2 mm high, and throughly wetted with mercury. The last 5 mm of each end of the sample is cut off with a clean razor blade and the fresh surfaces are pressed onto the mercury wetted plates. Such joints resemble solder joints and cannot be dismantled without cutting or otherwise destroying the sample. The combination of the brass plates and the K-Hg alloy produces high-resistance joints as is required to minimize current jetting.[6,12,13]

As usual, all sample preparation and handling was carried out in a glove box under cleaned argon. To facilitate mounting in a horizontal position, the samples rested on a Teflon plate which had most of the surface machined away to leave only occasional contact between the sample and plate. The heater and sample were constrained (relatively loosely) to lie along the direction of the magnetic field, but there was no impediment to relative thermal contraction between the Teflon plate and sample. Within a few minutes after mounting, the samples had begun the slow cooling to liquid nitrogen temperatures; annealing was not attempted because of the possibility of Hg diffusion into the central section of the sample. In the determination of γ_{zz}, temperature differences ΔT were typically 5–30 mK, necessitating a very careful handling of the magnetoresistance corrections to the carbon thermometers. Some measure of confidence in the corrections is gained from the simple check of measuring the mean sample temperature T and the difference ΔT and between the limbs at zero heat current and constant bath temperature (i.e., fixed T and zero ΔT) over the magnetic field range of 0–7.5 T. Data uncorrected for the magnetoresistance give a change in T of up to ~70 mK; with the corrections T remains constant to $\lesssim 5$ mK and the ΔT are zero to $\lesssim 0.5$ mK.

I have reliable data on seven samples, all with residual resistance ratios ($\rho_{293}/\rho_{1.4}$) in the range $(8.2–12)\times 10^3$. Although all the samples show saturation effects, the field required for saturation varies over a relatively wide range; Figs. 1–3 show the range of behavior that I have encountered. I have plotted $\gamma_{zz}L_0T$ (where L_0 is the Sommerfeld value of the Lorenz number and T the temperature) for reasons that will become clear later. The samples showing the most pronounced saturation were samples 3 (Fig. 2), 4 and 5 (not shown), and 6 (Fig. 3), and were

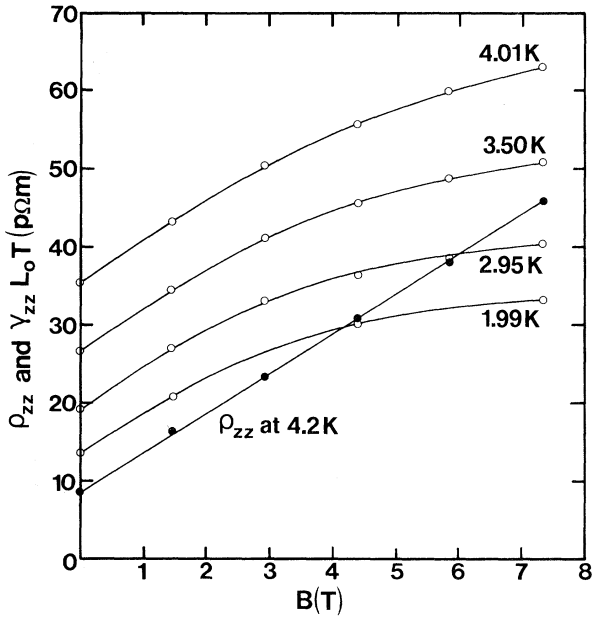

FIG. 1. The longitudinal electrical (ρ_{zz}) and thermal (γ_{zz}) resistivities of sample 2, the latter multiplied by L_0T, as a function of field B. This sample was oil coated.

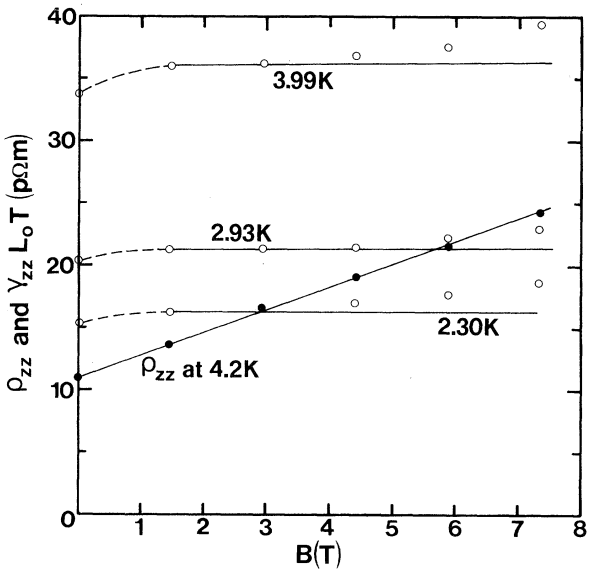

FIG. 2. Same as Fig. 1 for sample 3. This sample was dried with xylene.

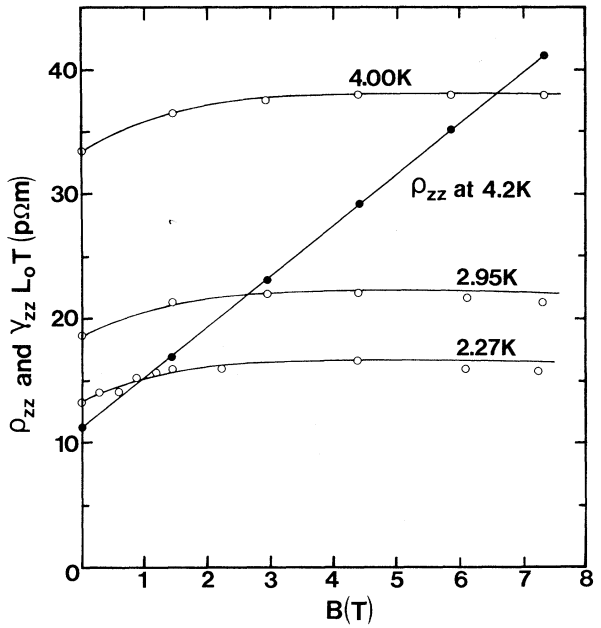

FIG. 3. Same as Fig. 1 for sample 6. This sample was dried with xylene.

those which were most carefully handled during mounting, were free of surface oil, and were aligned within about 2 deg of the field direction. Sample 3 (Fig. 2) shows a weak residual increase of γ_{zz} with field. This is thought to be due to a slight misalignment of sample and field. Sample 1 (not shown) was misaligned by 3–4 deg and exhibited much larger nonsaturating components.

Samples 1, 2, and 7 required higher fields for saturation. The results for sample 2 (Fig. 1) are typical of this group. This difference in behavior correlates with the fact that these samples were known to be damaged in various ways. Samples 1 and 2 were oil coated, which presumably causes internal damage due to differential thermal contraction, and sample 7 was intentionally stretched after mounting (and just before cooldown). It is possible that the absence of saturation of γ_{zz} that was indicated by the Tausch, Newrock, and Mitchel data[14] may have been caused by the use of samples with low λ_g in combination with the restriction of temperature range to $T \geq 3$ K; indeed their own data on λ_g are consistent with this hypothesis. It is very noticeable for this group of samples that the lower-field slopes of ρ_{zz} and $\gamma_{zz} L_0 T$ versus B are parallel and independent of T within experimental error. (See also the data of Ref. 14.) The void models predict this parallelism when the same electronic relaxation time is appropriate to both the thermal and electrical coefficients[9,10] i.e., $\tau_\gamma = \tau_\rho$, and elementary arguments suggest that $\Delta\rho = \Delta\gamma L_0 T = \alpha A f \omega$ even when $\tau_\gamma \neq \tau_\rho$ [where A is defined from $\rho_0 = A\tau_\rho^{-1} = A(L_0 \times T\tau_\gamma)^{-1}$].

I have attempted a quantitative fit of B_s and $(\Delta\gamma/\gamma_0)_s$ for the undamaged samples by taking average data on λ_g from other experiments.[11] Using $(\Delta\gamma/\gamma_0)_s = \alpha f(\lambda_0/\lambda_g)^{1/2}$ I have estimated αf from $\alpha f = \Delta\rho/\rho_0\omega\tau_\rho$, taking $\omega\tau_\rho = RB/\rho_0$, R being the Hall coefficient. To obtain B_s I assume $\omega\tau_\gamma = RB/\gamma_0 L_0 T$ and find $B_s = (L_0 T/R)(\gamma_0/\lambda_g)^{1/2}$. Table I lists the expected and observed values of B_s and $(\Delta\gamma/\gamma_0)_s$ for samples 3, 4, 5, and 6; although we do not find good agreement in each case, the observed magnitudes are generally of the correct order. Presumably the damaged samples have lower values of λ_g by virtue of dislocation scattering[11] and this will increase B_s and $(\Delta\gamma_{zz}/\gamma_0)_s$ but we have no quantitative way of estimating this effect at the present time.

To summarize, the predictions of the void models[6-10] are in qualitative agreement with the available experimental data on both the electrical and thermal resistivities of both compensated

TABLE I. Expected and observed values of B_s and $(\Delta\gamma/\gamma_0)_s$.

Sample	$10^{-3} \times (\rho_{293}/\rho_{1.4})$	T (K)	$(\Delta\gamma/\gamma_0)_s$ Expt.	$(\Delta\gamma/\gamma_0)_s$ Theory	B_s (T) Expt.	B_s (T) Theory
3	8.2	3.99	0.07	0.04	< 1.4	1.1
		2.30	0.07	0.06	< 1.4	0.7
4	11.9	3.97	0.2	0.4	~1	1.0
		2.34	0.4	0.9	~1	0.6
5	9.3	4.00	0.05	0.3	< 1.4	1.0
		2.27	0.15	0.5	< 1.4	0.6
6	8.5	4.00	0.14	0.10	1.7	1.0
		2.27	0.24	0.14	0.8	0.6

and uncompensated metals. To my knowledge, no other theory has been capable of predicting the observed diversity of behavior for the various coefficients of the two types of metals. Although I have been able to obtain reasonable quantitative agreement for some of the features of the present results, the fundamental parameter in the theory is f which is typically required to be $10^{-3} - 10^{-2}$, a range known to be too high.[15] This suggests that the theory is still in need of modification and improvement.

Dr. M. R. Stinson made available all his results before publication and provided many helpful comments concerning them. This work was supported by the National Sciences and Engineering Research Council of Canada.

[1]H. Taub, R. L. Schmidt, B. W. Maxfield, and R. Bowers, Phys. Rev. B 4, 1134 (1970).
[2]T. Amundsen and P. Jerstad, J. Low Temp. Phys. 15, 459 (1974).
[3]J. C. Garland and R. Bowers, Phys. Rev. 188, 1121 (1969).
[4]I. M. Lifshitz, M. Ya. Asbel', and M. I. Kaganov, Zh. Eskp. Teor. Fiz. 31, 63 (1956) [Sov. Phys. JETP 4, 41 (1957)].
[5]R. Fletcher, Solid State Commun. 21, 1139 (1977).
[6]J. S. Lass, J. Phys. C 3, 1926 (1970).
[7]J. B. Sampsell and J. C. Garland, Phys. Rev. B 13, 583 (1976).
[8]D. Stroud and F. P. Pan, Phys. Rev. B 13, 1434 (1976), and 20, 455 (1979).
[9]M. R. Stinson, Ph.D thesis, Queen's University, 1979 (unpublished), and J. Phys. F 10, L133 (1980).
[10]F. P. Esposito, R. S. Newrock, and K. Loeffler, Phys. Rev. B 20, 2348 (1979).
[11]M. R. Stinson, R. Fletcher, and C. R. Leavens, Phys. Rev. B 20, 3970 (1979).
[12]J. A. Delaney and A. B. Pippard, Rep. Prog. Phys. 35, 677 (1972).
[13]W. A. Reed, E. I. Blount, J. A. Marcus, and A. J. Arko, J. Appl. Phys. 42, 5453 (1971).
[14]P. J. Tausch, R. S. Newrock, and W. Mitchel, Phys. Rev. B 20, 501 (1979).
[15]D. R. Schouten and C. A. Swenson, Phys. Rev. B 10, 2175 (1974).

Dynamical Correlation Effects on the Quasiparticle Bloch States of a Covalent Crystal

G. Strinati,[a] H. J. Mattausch, and W. Hanke
Max-Planck-Institut für Festkörperforschung, 7000 Stuttgart 80, Federal Republic of Germany
(Received 9 June 1980)

A local-orbital formulation of the Dyson equation is presented for the one-particle Green's function with the self-energy expressed in terms of a dynamically screened interaction which includes local-field and excitonic effects. Also presented is a quantitative calculation of the quasiparticle states for diamond from first principles, which demonstrates the different roles played by two-particle excitations (electron-hole, plasmon). A discussion of the implications for local-density and empirical one-electron concepts is given.

PACS numbers: 71.45.Gm, 71.25.Cx, 71.25.Tn

The enormous success of energy-band calculations, using schemes such as the local-density approximation (LDA)[1] or the empirical (pseudo-) potential concept,[2] has given a kind of pragmatic answer of how to treat exchange and correlation effects on the one-electron states. On the other hand, the many-body theory of the solid state has established a number of more formal results, which may be viewed as a justification and first-principles basis for a one-electron theory.[1,3-6] For metals, in particular for the idealized homogeneous electron-gas model, a large amount of convergence of the two directions has been achieved.[1,3-6] In semiconductors and insulators this is not so, basically because of the strong localization and inhomogeneity of the electronic states. Some of the difficulties are revealed by a comparison of recent LDA calculations[7,8] with experimental gaps and valence-band widths, which show in both diamond (Table I) and silicon [$E_{\mathrm{gap}}^{\mathrm{LDA}} = 2.5$ eV vs $E_{\mathrm{gap}}^{\mathrm{exp}} = 3.4$ eV (Refs. 8, 9)] significant deviations. Empirical potentials, on the other hand, contain in a somewhat arbitrary manner higher-order [electron-hole (e-h), random-field approximation (RPA) local field] interaction effects.[10,11]

