Thermal magnetoresistance of potassium

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It has recently been shown that an inhomogeneous, anisotropic Hall coefficient, arising from a charge-density-wave domain structure, explains the nonsaturating electrical magnetoresistance of potassium. It is shown here that the same mechanism also explains the observed behavior of the thermal magnetoresistance. The transverse thermal magnetoresistance of a domain structure increases with increasing field, having both a linear and quadratic component. The longitudinal thermal magnetoresistance of a domain structure initially increases linearly with increasing field. Its behavior in higher fields, however, depends on whether or not the domain distribution is symmetric about the field direction. If the distribution is symmetric, saturation occurs; otherwise, a residual increase is possible.

I. INTRODUCTION

The nonsaturating electrical magnetoresistance of potassium has been a long-standing puzzle.[1] All workers have found that both the transverse[2] and longitudinal[3] electrical magnetoresistance increase linearly with increasing field, showing no sign of saturation in the highest magnetic fields ($B \sim 100$ kG). Fear that this was an artifact caused by probes led to the development of inductive techniques,[4] which confirm the results of probe methods.

It has recently been shown[5] that the observed behavior of the electrical magnetoresistance is explained by an inhomogeneous, anisotropic Hall coefficient, arising from a charge-density-wave domain structure.[6] The spatial variation of the Hall electric field distorts the current paths, causing both the transverse and longitudinal electrical magnetoresistance to increase. The Kohler slopes, calculated by effective-medium theory, agree with observed values.

The next major test of this theory is whether it can also explain the observed behavior of the thermal magnetoresistance.[7] The transverse thermal magnetoresistance of potassium increases with increasing field, having both a linear and quadratic component.[8–11] The longitudinal thermal magnetoresistance of potassium initially increases linearly with increasing field,[10] but saturates in higher fields.[12]

In high fields the thermal magnetoresistance behaves differently than the electrical magnetoresistance. This difference is caused by the lattice thermal conductivity. Whereas the electric current is carried only by electrons, the heat current is carried by both electrons and phonons. In unstrained, high-purity potassium at low temperatures, the ratio of the lattice to electronic thermal conductivity is typically 10^{-3}.[13] Although the lattice thermal conductivity is therefore negligible in zero magnetic field, it becomes significant in high fields, since the electronic thermal conductivity is reduced by a magnetic field.

The theory of the electrical magnetoresistance is easily extended to the thermal magnetoresistance by assuming an anisotropic Righi-Leduc (thermal Hall) coefficient. (Although the measured Righi-Leduc coefficient in potassium is approximately equal to the free-electron value,[11,14] its anisotropy has not been investigated.) It will be shown that, when this assumption for the electronic thermal conductivity is combined with the lattice thermal conductivity, the observed behavior of the thermal magnetoresistance is explained.

II. ANISOTROPIC RIGHI-LEDUC COEFFICIENT

The new ingredient of the theory for the electrical magnetoresistance was the discovery of the Hall-resistivity relation in potassium.[15] The Hall coefficient is the ratio of the transverse electric field E_H to the product jB of the current density and the applied magnetic field. In the usual transverse Hall effect, **B** is perpendicular to **j**; in the longitudinal Hall effect, **B** is parallel to **j**. It was found that both the transverse and longitudinal Hall coefficients of potassium are anisotropic, depending on the angle θ between **B** and the charge-density-wave (CDW) wave vector **Q**. This explained the amplitude and phase of the four-peak induced-torque patterns, observed in single-crystal spheres of potassium.[16–19]

The electrical magnetoresistivity of a **Q** domain, required to explain the induced-torque anomalies, is

$$\mathbf{E}=\rho[\mathbf{j}+t\omega_c\tau\mathbf{j}\times\hat{\mathbf{B}}+\alpha t\omega_c\tau(\hat{\mathbf{Q}}\cdot\hat{\mathbf{B}})\mathbf{j}\times\hat{\mathbf{Q}}] \ . \tag{1}$$

$\rho=m/ne^2\tau$ is the zero-field resistivity, $\omega_c=eB/mc$ is the cyclotron frequency, and τ is the electron relaxation time. t is the ratio of the Hall coefficient to the free-electron value, when **B** and $\hat{\mathbf{Q}}$ are perpendicular, and α is the Hall-coefficient anisotropy. t is approximately equal to 1.0 and α is at least as large as 0.3; $t=1.0$ and $\alpha=0.3$ are therefore assumed. For simplicity, the zero-field resistivity anisotropy[20] and open-orbit magnetoresistance[21] have been omitted.

At sufficiently low temperatures when electron scattering is elastic, the Wiedemann-Franz law[22] holds: The electronic thermal conductivity κ_e is proportional to the

trical conductivity σ,

$$\kappa_e = LT\sigma \ , \tag{2}$$

$L = 2.44 \times 10^{-8}$ W Ω/K^2 being the Sommerfeld value of the Lorenz ratio. This holds even in a magnetic field. However, already at 4 K in potassium there is enough inelastic scattering that the Wiedemann-Franz law is not quantitatively accurate.[23] Consequently, it is assumed that κ_e and σ still have the same magnetic-field dependence, but $L = L(T)$ in (2) is allowed to be temperature dependent. Accordingly, the electronic thermal magnetoresistivity of a Q domain is

$$\nabla T = \gamma_e [\mathbf{j} + t\omega_c\tau \mathbf{j}\times\hat{\mathbf{B}} + \alpha t\omega_c\tau(\hat{\mathbf{Q}}\cdot\hat{\mathbf{B}})\mathbf{j}\times\hat{\mathbf{Q}}] \ , \tag{3}$$

γ_e being the zero-field electronic thermal resistivity.

III. THERMAL MAGNETORESISTANCE OF A Q DOMAIN

It is instructive to derive first the thermal magnetoresistance of a free-electron metal.[24] In a magnetic field $\mathbf{B} = B\hat{\mathbf{z}}$, the electronic thermal conductivity is

$$\overleftrightarrow{\kappa}_e = \kappa_e \begin{bmatrix} \dfrac{1}{1+(\omega_c\tau)^2} & \dfrac{-\omega_c\tau}{1+(\omega_c\tau)^2} & 0 \\ \dfrac{\omega_c\tau}{1+(\omega_c\tau)^2} & \dfrac{1}{1+(\omega_c\tau)^2} & 0 \\ 0 & 0 & 1 \end{bmatrix} . \tag{4}$$

The lattice thermal conductivity is isotropic and field independent,

$$\overleftrightarrow{\kappa}_g = \begin{bmatrix} \kappa_g & 0 & 0 \\ 0 & \kappa_g & 0 \\ 0 & 0 & \kappa_g \end{bmatrix} . \tag{5}$$

The total thermal conductivity is then the sum

$$\overleftrightarrow{\kappa} = \overleftrightarrow{\kappa}_e + \overleftrightarrow{\kappa}_g \ . \tag{6}$$

Inverting (6) yields the transverse and longitudinal thermal magnetoresistance,

$$\Delta\gamma_{xx} = \Delta\gamma_{yy} = \frac{\gamma\lambda(\omega_c\tau)^2}{(1+\lambda)^2+\lambda^2(\omega_c\tau)^2} \ , \tag{7}$$

$$\Delta\gamma_{zz} = 0 \ , \tag{8}$$

and the Righi-Leduc coefficient $R_L \equiv \gamma_{yx}/B$,

$$R_L = \frac{R_L^{(0)}}{(1+\lambda)^2+\lambda^2(\omega_c\tau)^2} \ . \tag{9}$$

$\lambda \equiv \kappa_g/\kappa_e$ is the zero-field ratio of the lattice and electronic thermal conductivity, $\gamma = (\kappa_e+\kappa_g)^{-1}$ is the zero-field thermal resistance, and $R_L^{(0)} = -\gamma_e\omega_c\tau/B$ is the Righi-Leduc coefficient when $\kappa_g = 0$. For $\lambda < 1$ and $\lambda(\omega_c\tau) < 1$,

$$\Delta\gamma_{xx} = \Delta\gamma_{yy} \simeq \gamma\lambda(\omega_c\tau)^2 \ . \tag{10}$$

Even for a free-electron metal, the transverse thermal magnetoresistance is nonsaturating, increasing quadratically. The longitudinal thermal magnetoresistance of a free-electron metal vanishes.

In order to derive the thermal magnetoresistance of a Q domain, we first obtain the electronic thermal conductivity. Substituting $R_L^{(0)} = -\gamma_e\omega_c\tau/B$ in (3) yields

$$\nabla T = \gamma_e \mathbf{j} - tR_L^{(0)}\mathbf{j}\times\mathbf{B} - \alpha tR_L^{(0)}(\hat{\mathbf{Q}}\cdot\mathbf{B})\mathbf{j}\times\hat{\mathbf{Q}} \ . \tag{11}$$

This may be written in the same form as the free-electron thermal magnetoresistivity,

$$\nabla T = \gamma_e \mathbf{j} - R_L^{(0)}\mathbf{j}\times\mathbf{B}' \tag{12}$$

by introducing an effective magnetic field

$$\mathbf{B}' \equiv t[\mathbf{B} + \alpha(\hat{\mathbf{Q}}\cdot\mathbf{B})\hat{\mathbf{Q}}] \ . \tag{13}$$

Consequently, in an $x'y'z'$ coordinate system with the z' axis parallel to $\mathbf{B}'$, the electronic thermal conductivity $\overleftrightarrow{\kappa}_e$ is the same for a Q domain as for free electrons, with $\omega_c = eB/mc$ in (4) replaced by $\omega_c' = eB'/mc$. From (13),

$$(\omega_c'\tau)^2 = (\omega_c\tau)^2 t^2[1+\alpha(2+\alpha)\cos^2\theta] \tag{14}$$

with $\hat{\mathbf{Q}}\cdot\hat{\mathbf{B}} = \cos\theta$.

Adding the electronic and lattice thermal conductivities and inverting the total yields the thermal magnetoresistivity. In an xyz coordinate system with the z axis parallel to $\mathbf{B}$ and the x axis in the plane formed by $\mathbf{B}$ and $\mathbf{Q}$, the transverse and longitudinal magnetoresistances are

$$\Delta\gamma_{xx} = \gamma\lambda(\omega_c\tau)^2 t^2(1+\alpha\cos^2\theta)^2/D \ , \tag{15}$$

$$\Delta\gamma_{yy} = \gamma\lambda(\omega_c\tau)^2 t^2[1+\alpha(2+\alpha)\cos^2\theta]/D \ , \tag{16}$$

$$\Delta\gamma_{zz} = \gamma\lambda(\omega_c\tau)^2 t^2\alpha^2\cos^2\theta\sin^2\theta/D \ , \tag{17}$$

and the Righi-Leduc coefficient is

$$R_L = R_L^{(0)} t(1+\alpha\cos^2\theta)/D \ , \tag{18}$$

where

$$D = (1+\lambda)^2 + \lambda^2(\omega_c\tau)^2 t^2[1+\alpha(2+\alpha)\cos^2\theta] \ . \tag{19}$$

θ is the angle between $\mathbf{Q}$ and $\mathbf{B}$, $\lambda \equiv \kappa_g/\kappa_e$ is the zero-field ratio of the lattice and electronic thermal conductivity, $\gamma = (\kappa_e+\kappa_g)^{-1}$ is the zero-field thermal resistance, and $R_L^{(0)} = -\gamma_e\omega_c\tau/B$ is the free-electron Righi-Leduc coefficient when $\kappa_g = 0$. For $\lambda < 1$ and $\lambda(\omega_c\tau) < 1$, $D \simeq 1$ so that

$$\Delta\gamma_{xx} \simeq \gamma\lambda(\omega_c\tau)^2 t^2(1+\alpha\cos^2\theta)^2 \ , \tag{20}$$

$$\Delta\gamma_{yy} \simeq \gamma\lambda(\omega_c\tau)^2 t^2[1+\alpha(2+\alpha)\cos^2\theta] \ , \tag{21}$$

$$\Delta\gamma_{zz} \simeq \gamma\lambda(\omega_c\tau)^2 t^2 a^2\cos^2\theta\sin^2\theta \ . \tag{22}$$

As a consequence of the longitudinal-transverse mixing in (3), both the longitudinal and transverse magnetoresistances of a Q domain are, in general, nonsaturating, increasing quadratically. The longitudinal thermal magnetoresistance is a maximum when $\theta = 45°$ and vanishes when $\theta = 0°$ or 90°.

IV. THERMAL MAGNETORESISTANCE OF A DOMAIN STRUCTURE

The CDW wave vector **Q** in potassium, determined by neutron diffraction,[25,26] is tilted 0.85° away from a [110] direction and lies in a plane rotated 47° away from the (001) plane. Because of the underlying cubic symmetry, there are 24 symmetry-related **Q** directions, grouped in clusters of four about the six [110] axes. A single crystal is expected to be divided into **Q** domains, each having its **Q** along one of the 24 preferred axes. Since the thermal magnetoresistivity of a **Q** domain depends on the angle between **Q** and **B**, the thermal magnetoresistivity of a single crystal is then inhomogeneous.

The effective thermal magnetoresistance of a domain structure is evaluated by means of the effective-medium approximation.[27–30] This is a mean-field theory, which is expected to be valid when the domain size is greater than both the electron and phonon mean-free path but smaller than the sample dimensions. For $T=1.5$ K in unstrained, high-purity potassium, the electron and phonon mean-free paths are both about 0.1 mm; the domain size may vary, but is estimated to be also about 0.1 mm.[21,31] Since the constitutive relations, conservation laws, and boundary conditions have the same form for electric and thermal currents, the equations for the effective thermal conductivity are the same as for the effective electric conductivity.[32]

The domain distribution is specified by the volume fraction f_n of each of the 24 **Q** domain types. If the distribution is random, each **Q** domain occurs with equal probability $f_n=\frac{1}{24}$. The domain distribution, however, is not necessarily random, but may be textured. An example of a textured distribution is

$$f_n=\tfrac{1}{24}\{1+\beta[\tfrac{3}{2}(\hat{\mathbf{Q}}\cdot\hat{\mathbf{T}})^2-\tfrac{1}{2}]\}\ . \tag{23}$$

$\hat{\mathbf{T}}$ is the texture axis, and β is a texture parameter lying between -1 and $+2$. $\beta>0$ corresponds to a prolate texture and $\beta<0$ to an oblate texture.

The most striking difference between the behavior of the electrical and thermal magnetoresistance of potassium, which requires explanation, is the saturation of the longitudinal thermal magnetoresistance.[12] (The quadratic component of the transverse thermal magnetoresistance is expected, since it occurs even for free electrons.[24]) In Fig. 1 the longitudinal thermal magnetoresistance of a random domain distribution is plotted for different values of the ratio κ_g/κ_e. Surprisingly, even though γ_{zz} increases quadratically for a single domain, it saturates for a random domain distribution. In Fig. 1 the magnetic field **B** is parallel to the [100] crystallographic direction; similar results are obtained for other field directions.

From Fig. 1, γ_{zz} saturates when $\omega_c\tau\sim(\kappa_e/\kappa_g)^{1/2}$. The reason why the change occurs then can be understood by comparing the lattice thermal conductivity (5) with the electronic thermal conductivity of a **Q** domain, which has the same form as the free-electron thermal conductivity (4) in an appropriately chosen coordinate system. When $\omega_c\tau\sim(\kappa_e/\kappa_g)^{1/2}$, the lattice thermal conductivity has the same magnitude as the electronic thermal conductivity, thus being large enough to affect the thermal resistance.

In Fig. 2 the longitudinal thermal magnetoresistance of

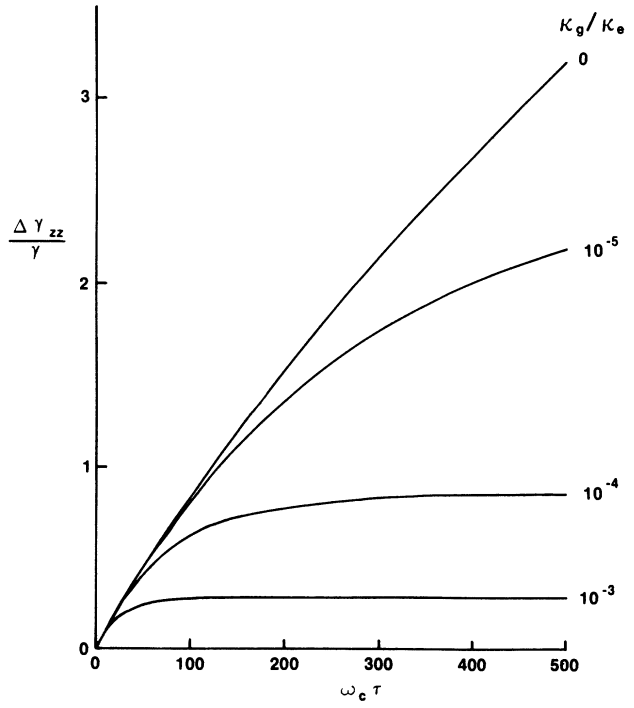


FIG. 1. Longitudinal thermal magnetoresistance of a random domain distribution. **B** is parallel to [100].

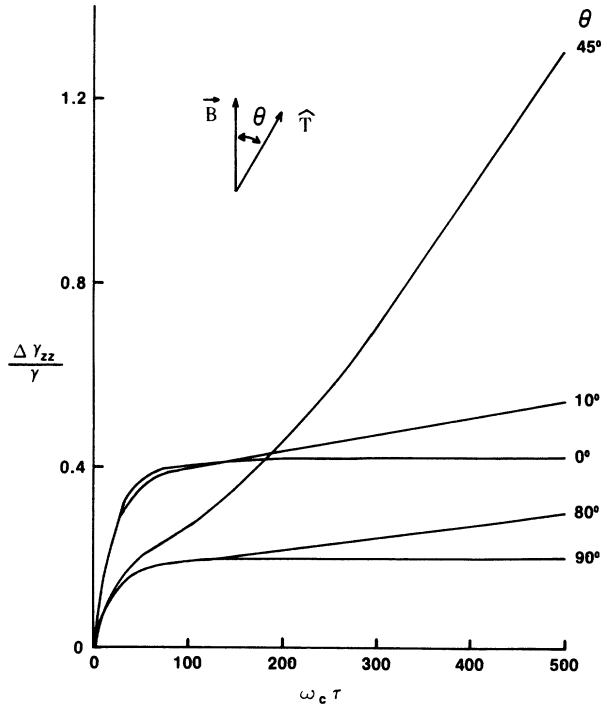
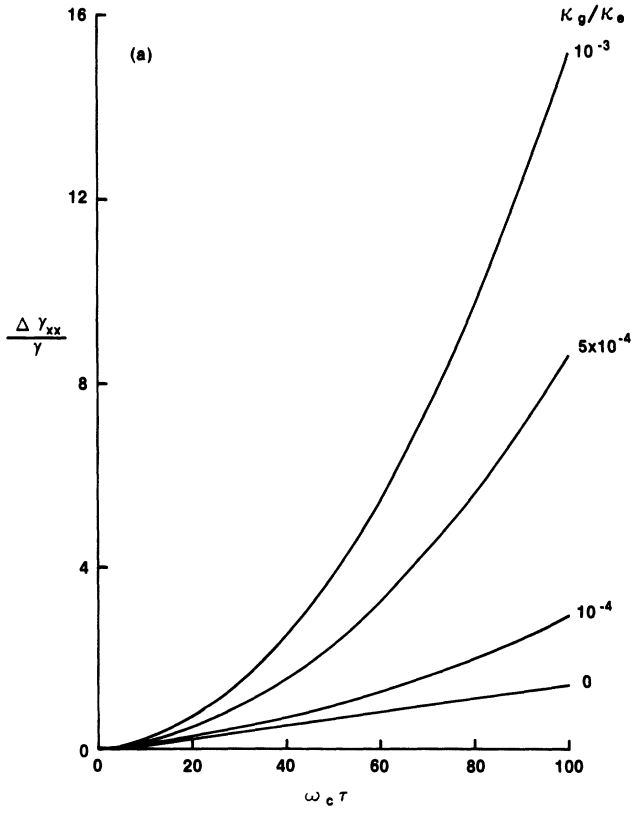


FIG. 2. Longitudinal thermal magnetoresistance of a textured domain distribution. $\kappa_g/\kappa_e=10^{-3}$. **B** is parallel to [100]. The texture axis $\hat{\mathbf{T}}$ lies in the (001) plane, making an angle θ with **B**; the texture parameter $\beta=+2$.



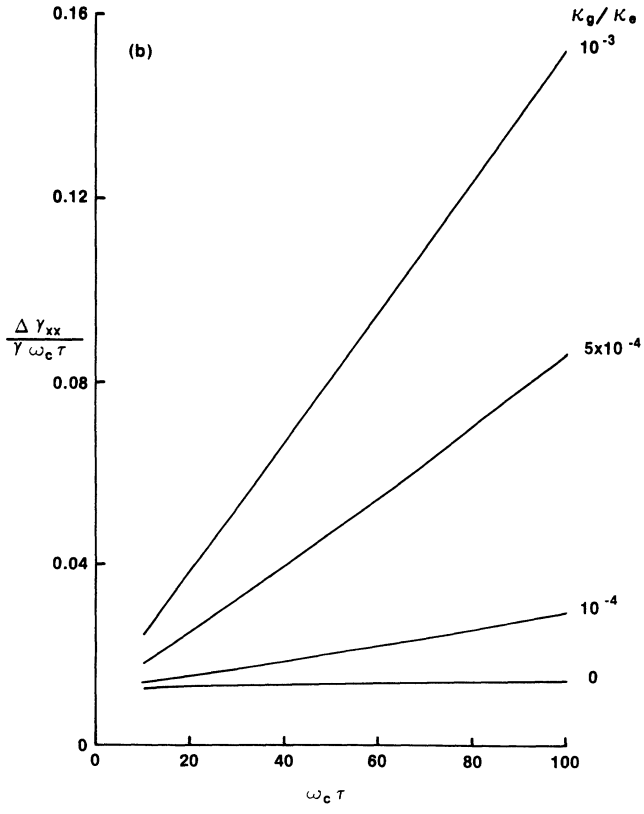


FIG. 3. Transverse thermal magnetoresistance of a random domain distribution. **B** is parallel to [100] and **j** is parallel to [010]. (a) $\Delta\gamma_{xx}/\gamma$ vs $\omega_c\tau$; (b) $\Delta\gamma_{xx}/\gamma\omega_c\tau$ vs $\omega_c\tau$.

a textured domain distribution is plotted for various angles θ between the texture axis and the applied magnetic field. For $\theta=0°$ or 90°, γ_{zz} saturates; otherwise, it continues to increase. Both prolate and oblate textures give similar results.

Such a residual increase has, in fact, been observed in some potassium samples.[12] It was attributed to a misalignment of the magnetic field with the current direction, since intentional misalignment by 3°–4° gave rise to much larger nonsaturating components. But, as illustrated in Fig. 2, the observed behavior could also have been caused by a textured domain distribution.

In Fig. 3(a) the transverse thermal magnetoresistance of a random domain distribution is plotted for different values of the ratio κ_g/κ_e. γ_{xx} is nonsaturating, having both a linear and quadratic component. This field dependence is shown more clearly in Fig. 3(b) by plotting $\Delta\gamma_{xx}/\omega_c\tau$ (for $\omega_c\tau>1$). Finally, in Fig. 4, the transverse thermal magnetoresistance of a textured domain distribution is plotted for various angles θ between the applied magnetic field and the texture axis, illustrating the effect of a preferred orientation on the magnitude of γ_{xx}.

V. CONCLUSION

It has been shown that an inhomogeneous, anisotropic Hall coefficient, arising from a charge-density-wave domain structure, explains the observed behavior of the

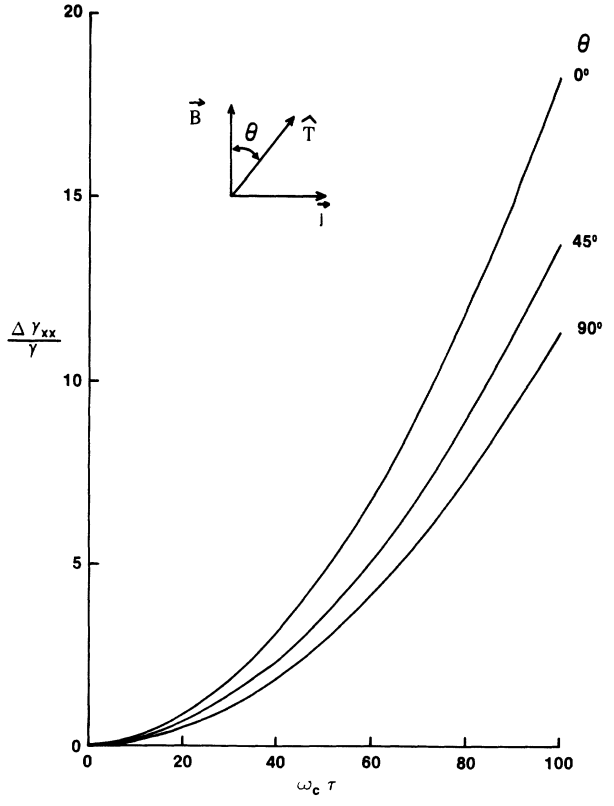


FIG. 4. Transverse thermal magnetoresistance of a textured domain distribution. $\kappa_g/\kappa_e=10^{-3}$. **B** is parallel to [100] and **j** is parallel to [010]. The texture axis $\hat{\mathbf{T}}$ lies in the (001) plane, making an angle θ with **B**; the texture parameter $\beta=+2$.

thermal magnetoresistance of potassium. (Obviously, the theory also applies to any other model of potassium which yields random anisotropies in the local Hall coefficient.) The only other theory which can explain both the electrical and thermal magnetoresistance of potassium is the void model,[33–38] but the void fraction required, which is of the order of several percent, is too large.[39,40]

A microscopic theory of the Hall-coefficient anisotropy, caused by a charge-density-wave structure, is still undeveloped. This remains a challenging project for future research.

ACKNOWLEDGMENT

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[32]See Ref. 5. The values of the eigenvalues Γ_i when $\lambda_1=\lambda_2=\lambda_3$ were unintentionally misstated there. The correct values are $\Gamma_1=\Gamma_2=\Gamma_3=-1/3\lambda_3$ when $\lambda_1=\lambda_2=\lambda_3$.

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