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Theoretical analysis of the transverse Hall effect in thin monocrystalline films

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The transverse Hall coefficient of thin monocrystalline films R_{HF} is derived from the recently presented bidimensional conduction model by introducing a term in the Boltzmann equation representing the effective mean free path. Numerical evaluations of R_{HF} show that the size effect in R_{HF} is less marked than that in resistivity and is much more sensitive to grain-boundary scattering than it is to external-surface scattering. Good agreement with the results from the previous experiments of several authors is found.

1. Introduction

The effect of external surfaces on the Hall coefficient R_{HF} of thin metal films subjected to a transverse magnetic field has been studied by many investigators [1–16], for the case of both polycrystalline and monocrystalline thin films. However, to our knowledge at present, no theoretical calculations have been undertaken of the conductivity and the Hall coefficient for a thin monocrystalline film placed in a transverse magnetic field when three types of electron scattering mechanisms are simultaneously operative, i.e. isotropic background scattering due to phonons and point defects, grain-boundary scattering and external-surface scattering.

In the absence of a magnetic field Mayadas and Shatzkes have proposed a conduction model [17] for monocrystalline films and polycrystalline films of constant grain size. However, this model is inadequate to describe the transport phenomena in presence of a transverse magnetic field because it assumes that only the grain-boundaries perpendicular to the applied electric field should be considered in calculations [17]; thus the Mayadas–Shatzkes model is, in practice, a one-dimensional model. Recently a study [18] has been devoted to a consideration of the theoretical electrical resistivity, due to the electron scattering both on external surfaces and on grain-boundaries; the grain-boundaries were represented by two series of planar potentials orientated respectively perpendicular to the x- and y-axes, the film surfaces being parallel to the plane determined by x- and y-axes.

In this paper an attempt is made to derive analytical expressions for the Hall coefficient R_{HF} and the conductivity σ_F of monocrystalline films whose grains exhibit a cubic shape by using a bidimensional conduction model [18] and by solving the Boltzmann equation determined using a mean free path method [19–21] under the application of a transverse magnetic field.

2. Theory

2.1. The effective relaxation time

In the absence of a magnetic field the transport properties of a thin monocrystalline film may be treated, to a good approximation, by a simple model [18] which states that in the case of a nearly-specular scattering on external surfaces ($p > 0.5$ where p is the specularity parameter) an effective mean free path, l_{eff}, may be defined which is given by

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$$l_{\text{eff}} = l_0 \left[1 + \frac{c^2}{\nu} - |\cos\theta| \cdot \left(\frac{c}{\nu} - \frac{1}{\mu}\right)\right]^{-1} \quad (1)$$

for the geometry shown in Fig. 1.

Thus the effective relaxation time, τ_{eff}, which described the effects of simultaneous background, grain-boundary and external-surface scatterings can be written as

$$\tau_{\text{eff}} = \frac{l_0}{v} \cdot \left[1 + \frac{c^2}{\nu} - |\cos\theta| \cdot \left(\frac{c}{\nu} - \frac{1}{\mu}\right)\right]^{-1},$$
$$= l_{\text{eff}} \cdot v^{-1} \quad (2)$$

where l_0 and v are, respectively, the background mean free path and electron velocity, c is a constant equal to $4/\pi$; ν and μ are related to grain size, a_g, transmission coefficient through grain-boundaries, t, film thickness a, specularity parameter, p, and mean free path, l_0, by equations:

$$\nu = \frac{a_g}{l_0 \cdot \ln\left(\frac{1}{t}\right)}; \quad (3)$$

$$\mu = \frac{a}{l_0 \cdot \ln\left(\frac{1}{p}\right)}. \quad (4)$$

2.2. Solving the Boltzmann equation

Consider a monocrystalline film with surfaces parallel to the x–y plane subjected to an electric field $(E_x, E_y, 0)$ in the plane of the film and to a transverse magnetic field $(0, 0, H)$ (Fig. 1); following similar lines to that of previous approaches [19–21], the appropriate Boltzmann equation can be written in the form:

$$\frac{f^1}{\tau_{\text{eff}}} - \frac{eH}{m}\left(v_y \frac{\partial f^1}{\partial v_x} - v_x \frac{\partial f^1}{\partial v_y}\right) = \frac{e}{m}\left(E_x \frac{\partial f^0}{\partial v_x} + E_y \frac{\partial f^0}{\partial v_y}\right), \quad (5)$$

where f^0 is the Fermi function and f^1 is the deviation of electron distribution f; $-e$ and v_x, v_y are the electron charge and the x- and y-components of the velocity v.

In order to solve the Boltzmann equation we put [19, 22]

$$f^1 = (v_x c_1 + v_y c_2) \frac{\partial f^0}{\partial v}, \quad (6)$$

where c_1 and c_2 do not depend explicitely on v_x and v_y and we introduce the complex quantities [17]:

$$g = c_1 - \mathrm{i}\, c_2; \quad (7)$$

$$F = E_x - \mathrm{i}\, E_y. \quad (8)$$

Then Equation 5 becomes

$$\frac{g}{\tau_{\text{eff}}} + \mathrm{i}\frac{v}{r} g = \frac{e}{mv} F. \quad (9)$$

It may be noted that the form of the effective mean free path (Equation 1) shows that the assumptions made about c_1 and c_2 are realistic moreover the above analysis supposes that $cg^{-1} - \mu^{-1}$ is not equal to zero.

Introducing the parameter $\alpha = l_0 r^{-1}$, where r is the radius of a free electron orbit in a magnetic field such that

$$r = \frac{mv}{eH}, \quad (10)$$

we then find for the general solution of Equation 9

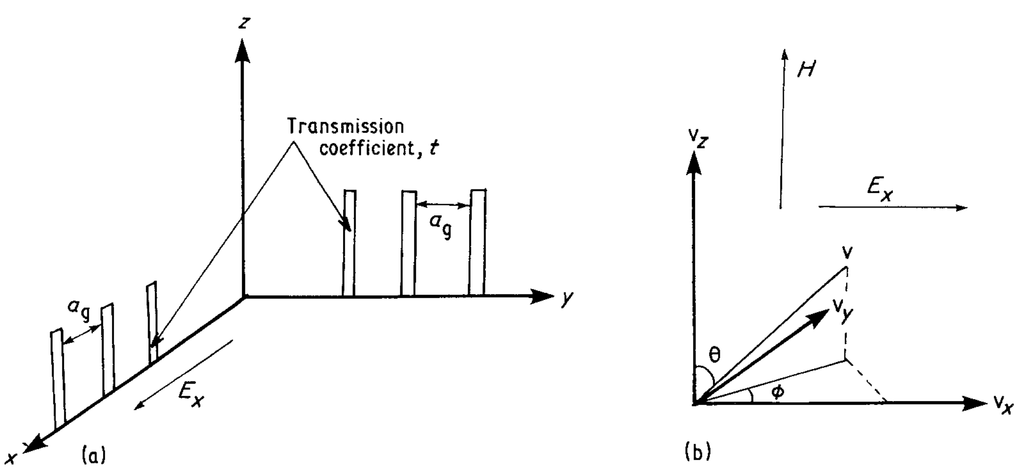

Figure 1 Geometry of the model.

$$g = \frac{e\,l_0}{m v^2} \cdot \frac{[E_x(\beta + b|\cos\theta|) - \alpha E_y] - \mathrm{i}\,[E_y(\beta + b|\cos\theta|) + \alpha E_x]}{[\beta + b|\cos\theta|]^2 + \alpha^2} \tag{11}$$

with

$$\beta = 1 + c^2 \nu^{-1} \tag{12}$$

and

$$b = \mu^{-1} - \nu^{-1}\,c. \tag{13}$$

2.3. The electrical conductivity

Introducing the polar co-ordinates (v, θ, ϕ) where $v_z = v\cos\theta$, the expressions for the total current densities in the x- and y-directions, J_x and J_y, can be written as

$$J_x = 2e\left(\frac{m}{h}\right)^3 v^4 \int_0^{2\pi} \cos^2\phi \; \mathrm{d}\theta \times \int_0^{\pi} c_1 \sin^3\theta\,\mathrm{d}\theta \tag{14}$$

and

$$J_y = 2e\left(\frac{m}{h}\right)^3 v^4 \int_0^{2\pi} \sin^2\phi \; \mathrm{d}\phi \times \int_0^{\pi} c_2 \sin^3\theta\,\mathrm{d}\theta. \tag{15}$$

Integration over θ and ϕ gives

$$J_x = \frac{3}{2}\sigma_0\,(A\cdot E_x - \alpha B\cdot E_y) \tag{16}$$

and

$$J_y = \frac{3}{2}\sigma_0\,(A\cdot E_y + \alpha B\cdot E_x) \tag{17}$$

with

$$A = \frac{1}{b}\left\{-\frac{1}{2} + \frac{\beta}{b} + \frac{\alpha^2 + b^2 - \beta^2}{2b^2} \times \ln\left(1 + \frac{b^2 + 2b\beta}{\alpha^2 + \beta^2}\right) - \frac{2\alpha\beta}{b^2}\arctan\frac{b\alpha}{\alpha^2 + \beta(\beta + b)}\right\} \tag{18}$$

and

$$B = \frac{1}{b}\left\{-\frac{1}{b} + \frac{\beta}{b^2}\ln\left(1 + \frac{b^2 + 2b\beta}{\alpha^2 + \beta^2}\right) + \frac{b^2 + \alpha^2 - \beta^2}{\alpha b^2}\arctan\frac{b\alpha}{\alpha^2 + \beta(\beta + b)}\right\}. \tag{19}$$

σ_0 is the background conductivity which is expressed as

$$\sigma_0 = \frac{n\,e^2\,l_0}{m\,v}. \tag{20}$$

The electrical conductivity, σ_F, of the monocrystalline film could be calculated according to the definition [22, 23]

$$\sigma_F = \left.\frac{J_x}{E_x}\right|_{J_y = 0}, \tag{21}$$

which yields

$$\frac{\sigma_F}{\sigma_0} = \frac{3}{2}\,\frac{A^2 + \alpha^2 B^2}{A}\;; b \neq 0. \tag{22}$$

2.4. The Hall coefficient, R_{HF}

The Hall coefficient of a thin monocrystalline film is defined by [22, 23]:

$$R_{HF} = \left.\frac{E_y}{H\cdot J_x}\right|_{J_y = 0.} \tag{23}$$

Equations 16 and 17 then give

$$R_{HF} = -\frac{2}{3}\,\frac{\alpha B}{\sigma_0\cdot H\cdot [A^2 + \alpha^2 B^2]}. \tag{24}$$

As it is well-known that in the free-electron model the Hall coefficient R_{HO} of the bulk metal is related to the number of free-electrons, n, by the following relation [3, 19, 22, 24]:

$$R_{HO} = -1/n\,e, \tag{25}$$

the ratio R_{HF}/R_{HO} of the Hall coefficient of a thin monocrystalline film to that of the bulk material may be written in the final form:

$$R_{HF}/R_{HO} = \frac{2}{3}\,\frac{B}{A^2 + \alpha^2 B^2}\;; b \neq 0. \tag{26}$$

2.5. The particular case of $b = 0$

For thin monocrystalline films of thicknesss such as to give

$$\mu = \nu/c \tag{27}$$

the expressions of the current densities reduce to

$$J_x = \frac{3}{4}\sigma_0 \int_0^{\pi} \frac{\beta E_x - \alpha E_y}{\beta^2 + \alpha^2}\sin^3\theta\,\mathrm{d}\theta \tag{28}$$

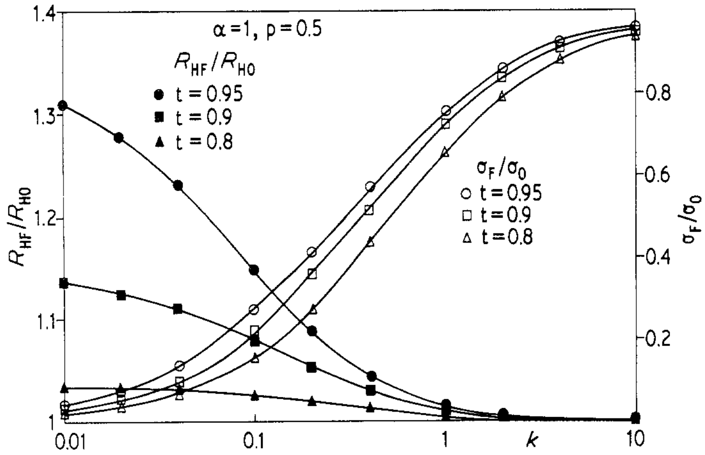

Figure 2 Variations in reduced Hall coefficient, R_{HF}/R_{HO}, and reduced conductivity, σ_F/σ_0, with reduced thickness, k, for $\alpha = 1, p = 0.5$ and a set of values for t.

and

$$J_y = \frac{3}{4}\sigma_0\int_0^{\pi} \frac{\beta E_y + \alpha E_x}{\beta^2 + \alpha^2} \sin^3\theta \, d\theta. \quad (29)$$

Hence,

$$\sigma_F/\sigma_0 = \beta^{-1}; \quad b = 0 \quad (30)$$

and

$$R_{HF}/R_{HO} = 1; b = 0. \quad (31)$$

Numerical values of R_{HF}/R_{HO} and σ_F/σ_0 can be evaluated with the aid of a pocket calculator. One main feature is shown by the variations of R_{HF}/R_{HO} and σ_F/σ_0 with reduced film thickness k (Figs 2 and 3): the reduced Hall coefficient is almost equal to 1 in a large range of values for k (error less than 10% for $p = 0.5$, $t \leqslant 0.9$ and $k \geqslant 0.1$).

It can then be predicted that no size effect in the Hall coefficient can be attributed to the limitation of the electronic mean free path by the monocrystalline film surfaces when the film thickness is larger than the half bulk mean free path. Furthermore, the size effect in conductivity is much more marked that the size effect in Hall coefficient for any thickness.

3. Discussion

This feature agrees with the experimental constant value of R_{HF} in Al monocrystalline films prepared by chemical reduction [11]; it is also in agreement with data related to evaporated films [25].

It has been generally reported that the Hall coefficient of Cu films is roughly thickness independent above a thickness of 20 nm [4], 30 nm [26] or 40 nm [9]. Kinbara *et al.* [4] reported that a marked deviation from the constant value of R_H occurred for very thin films (for thickness < 20 nm) and suggested that the origin of this deviation was caused by lattice defects, which is not in agreement with the negligible effect of grain boundaries. An alternative justification could be

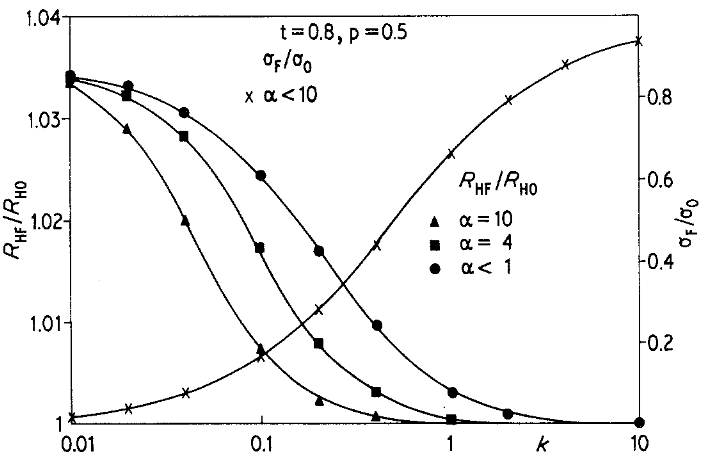

Figure 3 Variations in reduced Hall coefficient, R_{HF}/R_{HO}, and reduced conductivity, σ_F/σ_0, with reduced thickness, k, for $p = 0.5$, $t = 0.8$ and a set of values for α.

the existence of vacancies whose density is thickness-dependent; hence the size-dependence of resistivity and the deviation from the bulk value can be understood; the fact that irreversible changes occur in R_H and resistivity for very thin films left in the vacuum system could then be attributed to an apparent decrease in the density of carriers, which agrees with the observed [9] increase in resistivity but not with the observed increase in Hall coefficient.

In the case of antimony films [10] deposited on a substrate heated at 100 or 150° C, the variations in resistivity with thickness could correspond to a monocrystalline structure and the observed variations [10] in the Hall coefficient with thickness can be neglected from a statistical point of view.

The observed variations in resistivity with thickness of bismuth films [3, 5, 14] have a marked magnitude for film thicknesses larger than 100 nm and can be attributed to the usual size effect when assuming that the bulk mean free path takes a value near 1400 nm, no marked variation is observed in the Hall coefficient at room temperature [3, 5, 14, 27, 28], in accord with the predictions of the theoretical approach.

However in some cases [8, 28] a marked thickness-dependent of Hall coefficient is observed but experimental evidence has been given [8] for the existence of impurities in Cu films [8]; in the case of Bi films [28], it was suggested that grain-boundary scattering was the origin of the size effects; this assumption could agree with the above theoretical predictions since the experiments were performed at 4.2 K; nevertheless since Shubnikov–de Haas oscillations were observed [28] only qualitative agreement can be considered.

4. Conclusion

In the presence of a transverse magnetic field the Hall coefficient of thin monocrystalline films can be calculated from a bidimensional conduction model [18] in which an effective relaxation time describes the effects of simultaneous background, grain-boundary and external-surface scatterings. Theoretical expressions agree with experimental data on Cu, Ag, Sb and Bi thin films.

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