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Theoretical analysis of the Hall effect in thin polycrystalline metallic films

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In polycrystalline films where three types of scattering processes (background, grain-boundaries and external surfaces scatterings) are taking place at the same time an effective relaxation time is defined in the light of a three-dimensional model of grain-boundaries. Analytical expressions for the Hall coefficient and conductivity in thin polycrystalline metallic films subjected to a transverse magnetic field are then derived by using the Boltzmann transport equation. Previously published data can be theoretically interpreted in terms of the proposed model.

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 [1–11]. Measurements on polycrystalline or monocrystalline thin films have also been reported in the past few years [1, 2, 5, 7–10]. However, to our knowledge, there are at present no theoretical calculations of the conductivity and the Hall coefficient for a thin polycrystalline film placed in a transverse magnetic field in which 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.

Let us recall that Mayadas and Shatzkes (M–S) [12] have proposed a conduction model for polycrystalline films of constant grain size in the absence of a magnetic field. However, this model is inadequate to describe the transport phenomena in the presence of a transverse magnetic field because it assumes that only the grain-boundaries perpendicular to the applied electric field must be considered in the calculations [12], thus, the M–S model is in practice a one-dimensional model. For this purpose a previous work [13] has been devoted to theoretical electrical resistivity, due to the electron scattering both on external surfaces and on grain-boundaries; the grain-boundaries are represented by three series of potentials respectively oriented perpendicular to the x-, y- and z-axis.

In this paper an attempt is made to derive analytical expressions for the Hall coefficient R_{HF} and the conductivity σ_F of polycrystalline films whose grains exhibit a cuboid shape by using a three-dimensional conduction model [13] and by solving the Boltzmann equation determined in a mean free path method [14–16] 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 polycrystalline film may be treated to a good approximation by a simple model [13] which states that in the case of nearly specular scattering on external surfaces ($p \gtrsim 0.5$) an effective mean free path, l_{eff}, may be defined which is given by

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

for the geometry of Fig. 1.

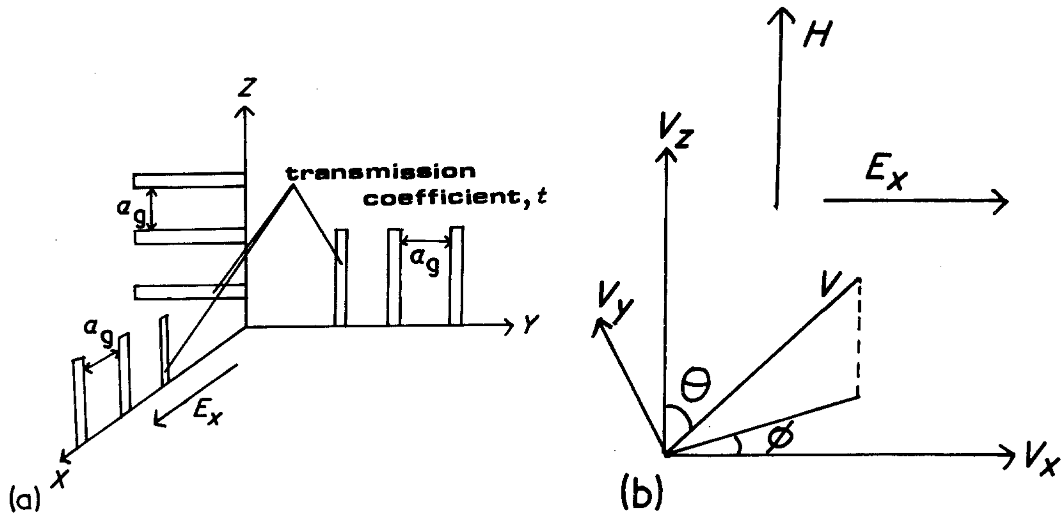

Figure 1 Geometry of the model; (a) the grain model and (b) the velocity co-ordinates.

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

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

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 the grain size a_g, transmission coefficient t through grain-boundaries, film thickness a, specularity parameter p and mean free path l_0 by Equations 3 and 4 below.

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

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

2.2. Solving the Boltzmann equation

Let us consider a polycrystalline 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 the lines of previous approaches [14–16], the appropriate Boltzmann equation can be written in the form

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

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

In order to solve the Boltzmann equation we can put [14, 17]

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

where c_1 and c_2 do not depend explicitly on v_x and v_y and the usual complex quantities are introduced [17]:

$$g = c_1 - (ic_2), \tag{7}$$

$$F = E_x - (iE_y). \tag{8}$$

Equation 5 then becomes

$$\frac{g}{\tau_{eff}} + i\frac{v}{r}g = \frac{e}{mv}F. \tag{9}$$

Note that on the one hand the form of the effective mean free path (Equation 1) indicates that the assumption made about c_1 and c_2 are reasonable and that on the other hand the above analysis supposes that $[(1-c)\cdot\nu^{-1}] + \mu^{-1}$ is not equal to zero.

The analysis involves a new quantity, r, namely the radius of a free electron orbit in a magnetic field

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

Introducing the parameter $\alpha = l_0 \cdot r^{-1}$ we then find for the general solution of Equation 6 that

$$g = \frac{e \cdot l_0}{mv^2} \times \left\{ \frac{[E_x(\beta + b\,|\cos\theta\,|) - \alpha E_y] - i\,[E_y(\beta + b\,|\cos\theta\,|) + \alpha E_x]}{[\beta + b\,|\cos\theta\,|]^2 + \alpha^2} \right\} \quad (11)$$

with

$$\beta = 1 + c^2 \cdot \nu^{-1} \quad (12)$$

and

$$b = \mu^{-1} + \nu^{-1} \cdot (1 - c). \quad (13)$$

2.3. The electrical conductivity

Introducing the polar co-ordinates (v, θ, ϕ) where $v_z = v \cdot \cos\theta$, we can at once write down the expressions for the total current densities in the x- and y-directions:

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

and (14)

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

Integration over θ gives

$$J_x = \tfrac{3}{2}\sigma_0(A \cdot E_x - \alpha B \cdot E_y) \quad (16)$$

and

$$J_y = \tfrac{3}{2}\sigma_0(A \cdot E_y + \alpha B \cdot E_x) \quad (17)$$

with

$$A = \frac{1}{b}\left\{ -\frac{1}{2} + \frac{\beta}{b} + \left[\frac{\alpha^2 + b^2 - \beta^2}{2b^2} \ln\left(1 + \frac{b^2 + 2b\beta}{\alpha^2 + \beta^2}\right)\right] - \left[\frac{2\alpha\beta}{b^2} \arctan\frac{b\alpha}{\alpha^2 + \beta(\beta + b)}\right]\right\}; \quad (18)$$

$$B = \frac{1}{b}\left\{ -\frac{1}{b} + \left[\frac{\beta}{b^2} \ln\left(1 + \frac{b^2 + 2b\beta}{\alpha^2 + \beta^2}\right)\right] + \left[\frac{b^2 + \alpha^2 - \beta^2}{\alpha b^2} \operatorname{arctang}\frac{b\alpha}{\alpha^2 + \beta(\beta + b)}\right]\right\}. \quad (19)$$

σ_0 is the background conductivity which is expressed as

$$\sigma_0 = \frac{ne^2 l_0}{mv}. \quad (20)$$

The electrical conductivity σ_F of the polycrystalline film could be calculated according to the usual definition [17, 18]

$$\sigma_F = \left.\frac{J_x}{E_x}\right|_{J_y = 0}. \quad (21)$$

This yields

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

2.4. The Hall coefficient, R_{HF}

The Hall coefficient of a thin film is defined by [17, 18]

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

Previous equations then give

$$R_{HF} = -\frac{2}{3}\left[\frac{\alpha B}{\sigma_0 \cdot H \cdot (A^2 + \alpha^2 B^2)}\right], \quad b \neq 0. \quad (24)$$

It is well-known that in the free-electron model the Hall coefficient R_{HO} of the bulk metal is related to the number, n, of free-electrons by the following relation [3, 14, 17, 19]:

$$R_{HO} = -1/ne. \quad (25)$$

The ratio R_{HF}/R_{HO} of the Hall coefficient of thin polycrystalline films to that of the bulk material may be written in the final form,

$$R_{HF}/R_{HO} = \frac{2}{3}\left(\frac{B}{A^2 + \alpha^2 B^2}\right), \quad b \neq 0. \quad (26)$$

2.5. The particular case, $b = 0$

For thin polycrystalline films of thickness as such as

$$\mu = \nu \cdot [c - 1]^{-1} \quad (27)$$

the expressions of the current densities reduce to

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

and

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

Hence

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

$$R_{HF}/R_{HO} = 1, \quad b = 0. \tag{31}$$

3. Discussion

Numerical values of the ratio R_{HF}/R_{HO} and σ_F/σ_0 may now be easily evaluated for different values of the parameters ν, μ and α with the aid of a pocket calculator.

3.1. The film conductivity

A further paper will report detailed results on the variations of the polycrystalline film conductivity and magnetoresistance with the physical parameters ν, μ and α. However, let us note that the theoretical plots of σ_F/σ_0 against μ for $\alpha = 0.1$ and for various values of ν (Fig. 2) show that the values of the ratio σ_F/σ_0 markedly depend on the grain parameter ν.

3.2. The Hall coefficient

First of all the limiting cases are examined where the effect of external surfaces (totally specular scattering or film of infinite thickness, a) or the effect of grain-boundaries (perfect transmission through grain-boundaries or grain of infinite size) can be neglected.

In the first case ($\mu \to \infty$), Equation 13 suggests that the problem is now formally identical to the problem (solved in a previous paper [20]) of a polycrystalline film in which only the background and grain-boundary scatterings are taken into account.

In the second case ($\nu \to \infty$) Equations 18 and 19 may be written in the limiting forms

$$A|_{\nu\to\infty} \approx \mu \left\{ -\frac{1}{2} + \mu + \frac{1}{2}(\alpha^2\mu^2 - \mu^2 + 1) \ln \left[\frac{\alpha^2 + (1+\mu^{-1})^2}{(1+\alpha^2)}\right] - 2\alpha\mu^2 \arctan \frac{\alpha}{\mu(\alpha^2 + 1 + \mu^{-1})} \right\} \tag{32}$$

and

$$B|_{\nu\to\infty} \approx \mu \left\{ -\mu + \mu^2 \ln \left[\frac{\alpha^2 + (1+\mu^{-1})^2}{(1+\alpha^2)}\right] + \left[\frac{\alpha^2\mu^2 - \mu^2 + 1}{\alpha} \arctan \frac{\alpha}{\mu(\alpha^2 + 1 + \mu^{-1})}\right]\right\}. \tag{33}$$

It ensures that in the absence of grain-boundary scattering effects, Equation 26 for the Hall coefficient reduces to that of a thin film which exhibits only the well-known Fuchs–Sondheimer size effect as previously derived [14].

Fig. 3 shows variations in the Hall coefficient ratio R_{HF}/R_{HO} plotted against reduced thickness k ($k = a/l_0$) for different values of the α parameter and for a given value of the grain parameter ($\nu = 1$) and specularity parameter ($p = 0.75$) while those of Fig. 4 show R_{HF}/R_{HO} as a function of k for five given values of ν and for a value of α ($\alpha = 0.1$) which corresponds to magnetic field magnitudes that are obtainable in practice [21].

In Fig. 5, plots of the ratio R_{HF}/R_{HO} against k

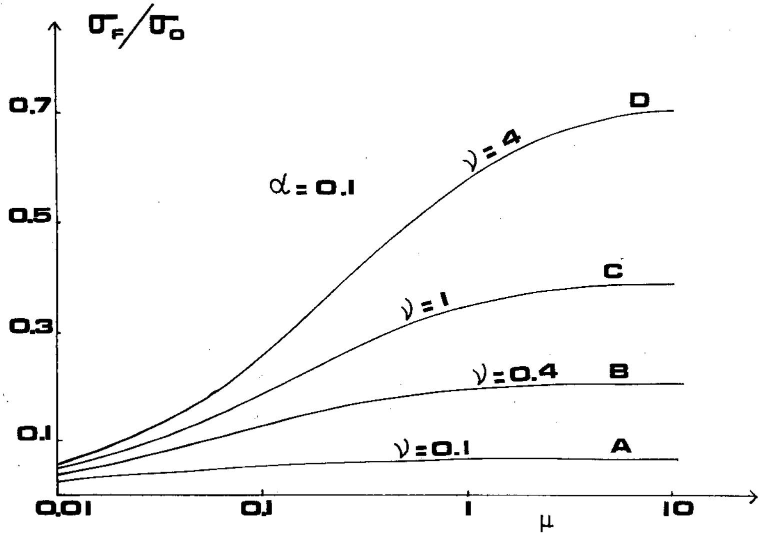

Figure 2 Variations in the conductivity ratio σ_F/σ_0 with μ (for $\alpha = 0.1$) under the influence of the grain parameter, ν. For curves A, B, C and D, ν = 0.1, 0.4, 1 and 4 respectively.

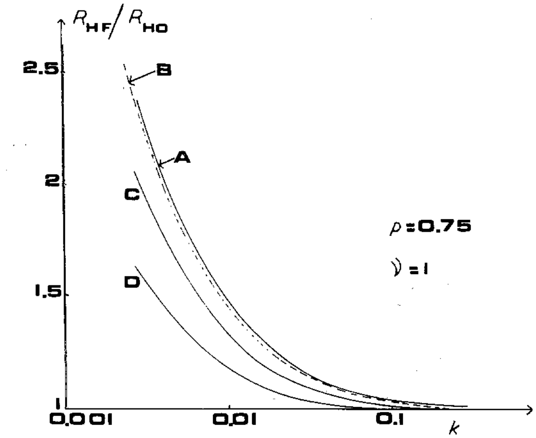

Figure 3 Variations in the Hall coefficient ratio R_{HF}/R_{HO} with the reduced thickness k (for $p = 0.75$ and $\nu = 1$) under the influence of the magnetic field, H. For curves A, B, C and D, $\alpha = 0.01$, 1, 4 and 10 respectively.

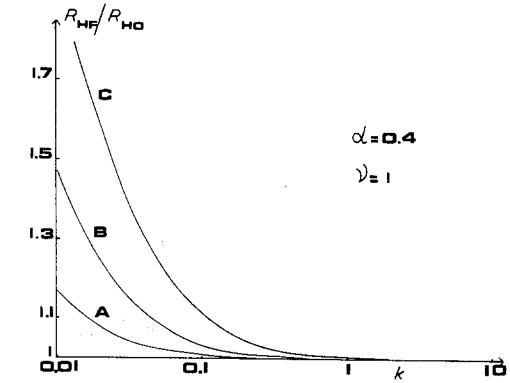

Figure 5 Variations in the Hall coefficient ratio R_{HF}/R_{HO} with the reduced thickness, k (for $\alpha = 0.4$ and $\nu = 1$) under the influence of the specularity parameter, p. For curves A, B and C, $p = 0.9$, 0.75 and 0.5 respectively.

are shown for various values of the specularity parameter p and for $\alpha = 0.4$ and $\nu = 1$.

Figs 3, 4 and 5 exhibit several features.

(a) For given values of α and ν, the curves have the usual aspects of the Sondheimer curves [17]. In particular it is noted that the physical requirement which states that R_{HF}/R_{HO} must decrease with increasing values of k and p is satisfied.

(b) For given values of p and α the variations in R_{HF}/R_{HO} with thickness markedly depend on the ν parameter. As the grain-boundary scattering effect becomes more significant, the ratio R_{HF}/R_{HO} tends to become insensitive to the reduced thickness variations at smaller thicknesses.

(c) In the limit of infinitely thick polycrystalline films and contrary to the electrical conductivity the ratio R_{HF}/R_{HO} is almost equal to unity. Hence, for polycrystalline films in which no size effects due to the geometrical limitation of the mean free path by the external surfaces occur, the changes of the Hall coefficient with the grain parameters (a_g, t) are negligible.

(d) For given values of k, p and ν the Hall coefficient ratio R_{HF}/R_{HO} increases (Fig. 5) with decreasing values of the α parameter, i.e. with decreasing values of the strength of the magnetic field. A similar feature has been observed by Li and Marsocci [22] and by Feder and Jossange [23] who have theoretically studied the variations of the Hall coefficient with the magnetic field strength using the framework of Sondheimer's theory [17].

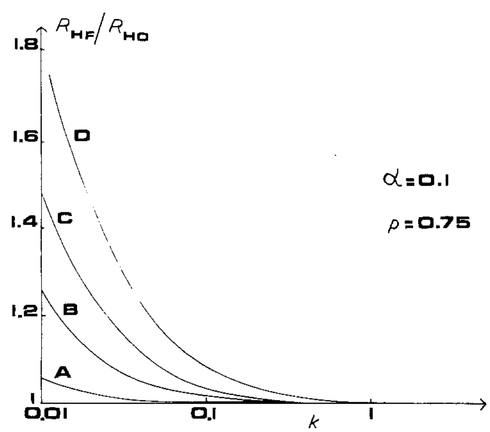

Figure 4 Variations in the Hall coefficient ratio R_{HF}/R_{HO} with the reduced thickness, k (for $p = 0.75$ and $\alpha = 0.1$) under the influence of the grain parameter, ν. For curves A, B, C and D, $\nu = 0.1$, 0.4, 1 and 4 respectively.

3.3. Comparison with experiments

Experiments [5, 8] on the thickness dependence of the Hall coefficient on thin polycrystalline films allow us to compare the observed results with the predictions of the proposed theory.

Simultaneous measurements of the film resistivity and Hall coefficient of polycrystalline copper films have been performed by Suri *et al.* [5]; it was observed that for thicker films, on the one hand that the limiting value of R_{H_∞} of the Hall coefficient ($R_{H_\infty} \approx 5.5 \times 10^{-5}\ cm^3\,C^{-1}$) does not significantly depart from the bulk value ($R_{HO} \approx 5 \times 10^{-5}\ cm^3\,C^{-1}$, [21]) and on the other hand that the deviation from the bulk value ρ_0 of the resistivity ρ_∞ of an infinitely copper film is more marked and depends on the deposition and annealing conditions [5]. For example, unannealed copper films exhibit deviations of about 60% for the resistivity and 7% for the Hall coefficient whereas departures of about 10% for the resistivity and 7% for the Hall coefficient are observed in the case of copper films annealed at 250° C.

Kinbara and Ueki [8] have reported similar results: for thick copper films R_{HF} was found to take a constant value of 6×10^{-5} cm^3 C^{-1} whereas the resistivity, ρ_∞, was found to be considerably larger ($\rho_\infty \approx 3\,\mu\Omega$ cm) than the bulk resistivity ($\rho_0 \approx 1.72\,\mu\Omega$ cm [24]).

These results may easily be understood both qualitatively and quantitatively in terms of the present theory [Section 3.2(c)]. It is reasonable to attribute the changes of ρ_∞ with annealing and deposition conditions to quantitative variations of grains, since it is well-known [25–32] that annealing induces a subsequent growth of grains; hence, from Equation 7, 12 and 13, the role played by the grains in determining ρ_∞ becomes less significant.

4. Conclusion

The three-dimensional model which assumes that the scattering effects of grain-boundaries can be described by three arrays of partially reflecting planes oriented perpendicular to the x-, y- and z-axes and that a single relaxation time can be defined for each type of scattering (background scattering, external surfaces, grain-boundary) can be considered as a convenient tool to derive an analytical expression for the Hall coefficient in thin polycrystalline films subjected to a transverse magnetic field. In marked contrast with the electrical conductivity, the Hall coefficient of sufficiently thick films is found to be independent of the grain parameters. Some experimental data on polycrystalline copper films can be easily interpreted in terms of the proposed model.

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