Size effects in metallic thin films

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The relaxation time derived with use of the thickness-dependent thin-film Thomas-Fermi impurity potential in the collision term of the Boltzmann transport equation is used to study the classical size effects in the Hall coefficient R_f and magnetoresistance M_f in metallic thin films. The theoretical prediction of R_f agrees well with experimental results. The dependence of M_f on thickness and magnetic field is also discussed.

A survey of the literature on the experimental and theoretical studies of metallic thin films[1–13] reveals that the transport properties, such as electrical resistivity ρ_f, magnetoresistance M_f, and the Hall coefficient R_f, show marked deviations from the bulk behavior, and the deviations become significant as the thickness becomes comparable with the mean free path of the electrons. In an earlier analysis[10] Ajoy, Devanathan, and Govindaraj (hereafter referred to as ADG) have shown the size-dependent behavior of ρ_f by using the energy, momentum, and thickness-dependent relaxation time τ. In the present work, we have extended the ADG theory to study the other transport quantities, viz., R_f and M_f, using τ. The Hall coefficient R_f shows a size effect, and it is found that R_f is large for small thickness, and decreases with increasing thickness until it reaches the bulk value R_b. Magnetic field shows a very small size effect in R_f. At low magnetic field, R_f is slightly higher than its bulk value, and approaches the bulk value R_b at strong magnetic fields. Theoretical predictions of R_f agree with the experimental data on potassium films. The size effect in M_f is observed as a function of thickness and magnetic field.

The energy, momentum, and thickness-dependent relaxation time in ADG is given by

$$\tau=\frac{Kd}{4\pi\lambda^2Nq_0^2F}\left(\frac{1}{\lambda^4}+\frac{1}{[\lambda^2+4(2E-K^2)]^2}\right)^{-1}, \quad (1)$$

where $F=[(L_b/2)^2+d^2]^{1/2}-d$, L_b is the bulk dimension, d is the thickness, $\lambda=6\pi n_0/E_F$, n_0 is the electron concentration, E_F is the Fermi energy, N is the concentration of the impurity atoms, q_0 is the charge of the impurity atom, K is the momentum, and E is the total energy of the electron. We use atomic units ($e^2=1$, $\hbar=1$, $m_e=1$) throughout this paper. Equation (1) serves as the basis for our analysis of the classical size effects in metallic thin films.

Expressions for the Hall coefficient R_f and the magnetoresistance M_f are derived for thin films using the relaxation time τ by suitably modifying the bulk expressions.[14] Expressions for the x and y components of the current density, J_x and J_y, for the metallic thin film subjected to an electric field E_{0x}, E_{0y}, in the plane of the film and transverse magnetic field **B** are given by

$$J_x=SE_{0x}+S^*E_{0y} \quad (2)$$

and

$$J_y=SE_{0y}-S^*E_{0x}, \quad (3)$$

where

$$S=-\sum_m\int d^2K\,\frac{\partial F_0}{\partial E}\frac{\tau}{1+\tau^2B^{*2}}K_j^2D_{\mathbf{K},m} \quad (4)$$

and

$$S^*=-\sum_m\int d^2K\,\frac{\partial F_0}{\partial E}\frac{\tau^2B^*}{1+\tau^2B^{*2}}K_j^2D_{\mathbf{K},m}. \quad (5)$$

τ is the relaxation time given by Eq. (1), $F_0=\psi^*_{\mathbf{K},m}(\mathbf{r})\psi_{\mathbf{K},m}(\mathbf{r})f_0(E)$, $\psi_{\mathbf{K},m}(\mathbf{r})$ is the wave function of the electron, and $f_0(E)$ is the Fermi-Dirac distribution function given by

$$f_0(E)=\{\exp[(E-E_F)/kT]+1\}^{-1}, \quad (6)$$

where $E=E_{\mathbf{K},m}$, E_F is the Fermi energy of the electron, K_j may be either the x or y component of K, $D_{\mathbf{K},m}$ is the density of states at $\mathbf{K},m$, and $\mathbf{B}^*=\mathbf{B}/c$. Substituting Eq. (1) in Eqs. (4) and (5) and evaluating the integrals, we get

$$S=\frac{1}{8\pi^2\lambda^2Nq_0^2F}\sum_{m=1}^{m_{\max}}A^{3/2}D^{-1}/H, \quad (7)$$

and

$$S^*=\frac{B^*d}{32\pi^2\lambda^4N^2q_0^4F^2}\sum_{m=1}^{m_{\max}}A^2D^{-2}/H, \quad (8)$$

where

$$A=[2E_F-(m\pi/d)^2],$$

$$D=\left(\frac{1}{\lambda^4}+\frac{1}{[\lambda^2+(2m\pi/d)^2]^2}\right),$$

$$H=[1+AG^2(\lambda^2FD)^{-2}] \text{ with } G=B^*d/4\pi N.$$

Equations (2), (3), (7), and (8) are used in the calculation of R_f and M_f.

The Hall coefficient is defined by

$$R_f=\left.\frac{E_{0y}}{BJ_x}\right|_{J_y=0}. \quad (9)$$

By combining Eqs. (2) and (3), we get

$$R_f = \frac{S^*}{B[S^2+(S^*)^2]} , \quad (10)$$

where S and S^* are given by Eqs. (7) and (8).

The magnetoresistance is defined by

$$M_f = \frac{\rho_{(B\neq 0)} - \rho_{(B=0)}}{\rho_{(B=0)}} , \quad (11)$$

where

$$\rho_{(B\neq 0)} = \left.\frac{E_{0x}}{J_x}\right|_{J_y=0} . \quad (11a)$$

Combining Eqs. (2) and (3), we get

$$\rho_{(B\neq 0)} = \frac{S}{S^2+(S^*)^2} . \quad (11b)$$

Therefore

$$M_f = \frac{SU}{S^2+(S^*)^2} - 1 , \quad (12)$$

where U is the electrical conductivity σ_f and is the same as in Eq. (37) in ADG

$$\sigma_f = \frac{1}{4\pi^2\lambda^2 N q_0^2 F}\frac{1}{2}\sum_{m=1}^{m_{\max}} A^{3/2}D^{-1} .$$

The quantities S^* and S are given by Eqs. (7) and (8).

The size effect in the Hall coefficient R_f is studied using Eq. (10). Figure 1 shows the thickness-dependent behavior of the Hall coefficient R_f for potassium thin films. The value of R_f is calculated in the presence of magnetic field $B=10^3$ G. Figure 1 clearly explains that the Hall coefficient is large for small thickness, and decreases with increasing thickness until it reaches the bulk value R_b at large thickness. The values of R_f are found to be in good agreement with the experimental results.[15] The value of R_f is also calculated for Cu and Ag thin films, and size effects in Cu and Ag are smaller than those for K at a given thickness. The field effect on R_f is studied, and it is found that for $d=100$ a.u., R_f remains almost constant up to 10^5 G, and for $d=1000$ a.u., a small decrease in R_f is observed at about 3×10^3 G and is just 1% at 10^5 G. The same effect is also observed for other metallic films. However, the variation of R_f with magnetic field is very small.

Equation (12) is used to study the dependence of magnetoresistance M_f upon thickness d and magnetic field B. The value of M_f is calculated for various values of d in the pres-

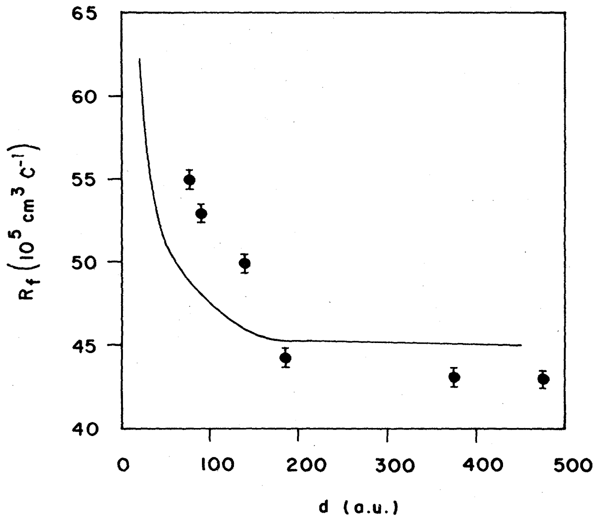

FIG. 1. The Hall coefficient as a function of thickness for potassium thin film. Experimental data on potassium thin film at 90 K (Ref. 15).

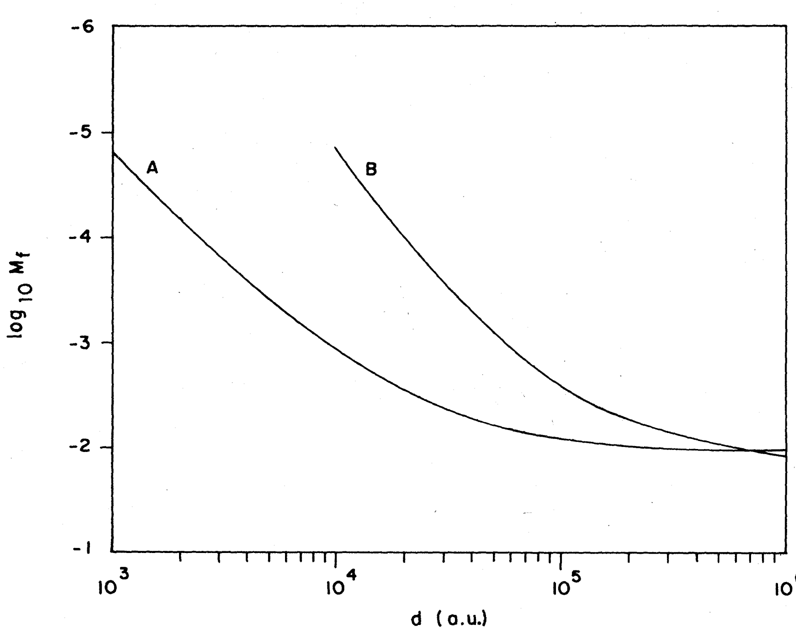

FIG. 2. Magnetoresistance as a function of thickness for copper thin films. Fur curve A, $B=10^3$ G, for curve B, $B=10^2$ G.

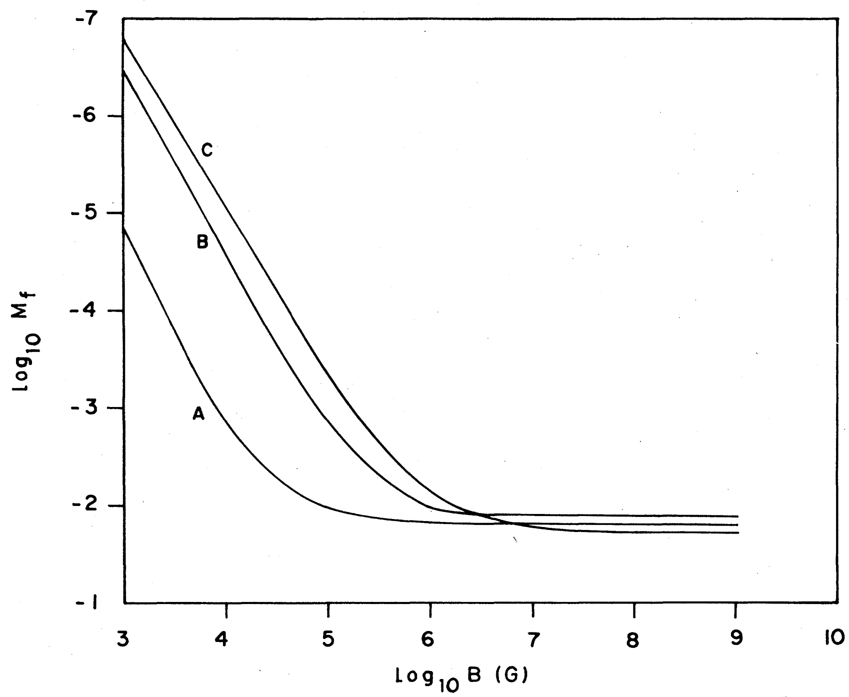

FIG. 3. Magnetoresistance as a function of magnetic field for copper thin films. For curve A, $d=1000$ a.u.; for curve B, $d=100$ a.u.; for curve C, $d=50$ a.u.

ence of magnetic field ($B=10^2$ and 10^3 G, respectively), and the dependence of $\log M_f$ on d is shown in Fig. 2. At small thickness, though, the value of M_f is very small, and the variation of M_f is large. At large thickness, the value of M_f is large, but its variation is small. The value of M_f approaches a constant value when the thickness reaches the bulk value. The value of M_f is calculated for $d=50$, 100, and 1000 a.u. in the presence of different values of magnetic field and the variation of $\log M_f$ with $\log B$ is plotted in Fig. 3. At low magnetic field, M_f is very small, but its variation with field is large. However at strong field, the value of M_f is large, and its variation with field is small. The value of M_f reaches a constant value when the field attains 10^7 G.

The present study on the Hall coefficient and magnetoresistance explains the existence of the size effect in metallic thin films as explained in other theories.[1,2,5] However, the present model does not include the specular parameter p for surface reflection, to explain the thickness dependence of R_f and M_f. The value of R_f is found to be larger than the bulk value, and the thickness dependence is observed down to 10^4 a.u. The thickness and magnetic field dependence features of M_f in the present model are similar to what has been observed in other works.[2,5,7]

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