TEMPERATURE DEPENDENCE OF THE HALL COEFFICIENT OF THIN FILMS

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A theoretical expression for the temperature coefficient $\beta_{RH}$ of the Hall coefficient $R_{HF}$ of metallic films is deduced from the Fuehs–Sondheimer conduction model. The general expression takes into account the deviation introduced by the geometrical limitation of the mean free path. This is negligible for relatively thick films ($k \gg 1$ for $p = 0$) and agrees with experiments previously reported by other authors.

1 INTRODUCTION

Many experimental papers have been published on the thickness and/or temperature dependence of the Hall coefficient of thin metallic films. Most of the experimental plots of the Hall coefficient, $R_{HF}$, as a function of the reduced thickness $k$ and/or temperature $T$ were related to bismuth films $^{1,3,6,8}$ and they were analyzed in terms of the quantum size effect $^{1-3}$ at low temperatures and used a two carrier model $^{1,3,6}$. However, on the one hand a number of transport properties (such as film resistivity $\rho_F$, temperature coefficient of resistivity $\beta_F$, $8-10$, Hall coefficient $R_H$ $^{10,11}$ have been compared with the predictions of the classical size effect $^{1-3}$ Fuchs–Sondheimer Theory). On the other hand, previously reported studies on silver films $^{5,9}$ have shown that a single carrier conductivity occurs (electronic conductivity) and that transport properties ($\rho_F$, $\beta_F$, $R_H$) exhibit a qualitative agreement with the Fuchs–Sondheimer theory (F–S theory) in the reduced thickness range $k$. Furthermore these authors have obtained linear $R_{HF}$ versus $T$ plots.

Hence, in the case of metal films for which the F–S assumptions are sufficient to explain most of the conduction phenomena, we investigated the temperature dependence of the Hall coefficient by considering the temperature dependence of two physical parameters, electron density $n$ and bulk mean free path $l_0$. Note that this study reports results on the variation of Hall coefficient with temperature $T$ in the relatively high temperature range $T \gg 100$ K.

2 THE TEMPERATURE DEPENDENCE OF THE HALL COEFFICIENT IN THE F–S MODEL

We have previously shown $^{14}$ that in the case of a low magnetic field (commonly the case observed in experimental works $^{1,2,5,8,10,12}$), the Hall coefficient $R_{HF}$ of thin metallic films in the presence of a transverse magnetic field can be represented as the product of the film resistivity $\rho_F$ and its temperature coefficient $\beta_F$

$$R_{HF}/R_{HO} \approx \frac{\rho_F \beta_F}{\rho_0 \beta_0}$$

(1)

where $R_{HO}$ the Hall coefficient of bulk metal is given by

$$R_{HO} = -\frac{1}{ne}$$

(2)

where $n$ is the number of free electrons per unit volume and $-e$ is the electronic charge.

In the F–S model the film resistivity $\rho_F$ is expressed as

$$\rho_F/\rho_0 = [1 - A(k)]^{-1}.$$  

(3)

We have previously $^{15}$ found that the film t.c.r. $\beta_F$ may be written in the forms

$$\beta_F/\beta_0 = 1 + \frac{k}{1 - A(k)} \frac{d[A(k)]}{dk} = 1 + \frac{-A(k) + B(k)}{1 - A(k)} = 1 + D(k)$$

(4)
where

\[ A(k) = \frac{3}{2k} \int_1^\infty \left[ \frac{1}{t^2} - \frac{1}{t^4} \right] \frac{1 - e^{-kt}}{1 - p e^{-kt}} dt \]

\[ B(k) = \frac{3}{2} \left[ 1 - p \right]^2 \int_1^\infty \left[ \frac{1}{t^2} - \frac{1}{t^4} \right] \frac{e^{-kt}}{[1 - p e^{-kt}]^2} dt \]

The reduced thickness \( k \) is the ratio of the film thickness \( a \) to the bulk mean free path \( l_0 \), i.e.

\[ k = a/l_0 \]  

The fraction of electrons specularly scattered on external surfaces is given by \( p \), \( \rho_0 \) is the resistivity of bulk metal and \( \beta_0 \) its temperature coefficient.

We immediately verify that, if we neglect (as generally assumed) the coefficients of linear expansion of the film, the Hall coefficient \( R_{HF} \) formula contains two parameters \( n \) and \( l_0 \) depending upon the temperature \( T \) so that

\[ R_{HF} = \phi \{ n(T), l_0(T) \} \]  

with

\[ \phi(n, l_0) = -\frac{1}{n e} \left[ 1 + D(k l_0) \right] \]  

Logarithmic differentiation of Eqs. 8 respect to \( l_0 \) and \( n \) gives

\[ \frac{dR_{HF}}{R_{HF}} = -\frac{dn}{n} \cdot \frac{d(1 + D(k))}{1 + D(k)} \cdot \frac{dl_0}{l_0} + \frac{d(1 - A(k))}{1 - A(k)} \frac{dl_0}{l_0} \]

Taking into account that with the above assumption

\[ \frac{dk}{k} = -\frac{dl_0}{l_0} \]

and then, calculating \( d(1 + D(k))/dl_0 \), we obtain, with the aid of Eq. (4)

\[ \frac{dR_{HF}}{R_{HF}} = -\frac{dn}{n} \left( \frac{dl_0}{l_0} \right) \left\{ \frac{D(k)}{1 + D(k)} \left[ (1 + C(k) + D(k)) \right] \right\} \]

\[ + D(k) \cdot \left( -\frac{dl_0}{l_0} \right) \]

where

\[ C(k) = \frac{k}{-A(k) + B(k)} \frac{dB(k)}{dk} \]

Defining the temperature coefficient \( \beta_{RH} \) of the Hall coefficient \( R_{HF} \) as:

\[ \beta_{RH} = \frac{1}{R_{HF}} \frac{dR_{HF}}{dT} \]

and the temperature coefficient \( \beta_n \) of the electronic density \( n \) as:

\[ \beta_n = -\frac{1}{n} \cdot \frac{dn}{dT} \]

and assuming that the Sommerfeld relation is valid (that leads to \( \rho_0 = -1/l_0 \frac{dl_0}{dT} \) we obtain:

\[ \beta_{n} = -\beta_n + \beta_0 \cdot f^*(k) \]

With

\[ f^*(k) = D(k) \left\{ 1 + \frac{1 + C(k) + D(k)}{1 + D(k)} \right\} \]

To determine the temperature coefficient \( \beta_{RH} \) we have to introduce an additional term which takes into account that the reduced thickness \( k = k(l_0) \) depends upon the temperature. As expected, this term \( \beta_0 \cdot f^*(k) \) describes the thickness variation of \( \beta_{RH} \).

In the next section we propose to evaluate the function \( f^*(k) \) and to demonstrate that generally the product \( \beta_0 \cdot f^*(k) \) becomes negligible in the limit of relatively large \( k \).

3 DISCUSSION

In the case of totally diffuse scattering on external surface Eq. (6) reduces to

\[ B(k) = \frac{3}{2} \int_1^\infty \left[ \frac{1}{t^2} - \frac{1}{t^4} \right] e^{-kt} dt \]

that leads to

\[ k \frac{dB(k)}{dk} = -\frac{3}{2} k \int_1^\infty \left[ \frac{1}{t^2} - \frac{1}{t^4} \right] e^{-kt} dt \]

Repeated integration by parts gives an alternative expression to Eq. (18) that can be numerically evaluated. The results of the calculation are shown in Figure 1 that illustrates the deviation \( f^*(k) \) introduced by the "size effect". The most interesting result is that the function \( f^*(k) \) decreases rapidly with increasing values of the reduced thickness and reaches zero as \( k \to 1 \). Then, even for relatively thin films the factor \( \beta_n \) is generally not appreciably affected by the product \( \beta_0 \cdot f^*(k) \) except if the value of \( \beta_n \) is considerably smaller than the \( \beta_0 \) value \((-\beta_n/\beta_0 < 10^{-1})\). A
fortiori we expect a similar behaviour when a partially specular scattering occurs on external surfaces. Now let us examine the experiments on silver films prepared by chemical reduction which have been carried out by Flechon, Drexler and Viard\textsuperscript{5,9}. These authors\textsuperscript{9} have compared their results on the thickness variation of the resistivity $\rho_F$ and its t.c.r. $\beta_F$ with the F–S theory. The agreement between theory and experiment is satisfactory and values of 0.5 and of about 400 Å are assigned to the specularly parameter $p$ and the mean free path of an infinitely thick silver film respectively. Further results on the Hall coefficient in the thickness range 500 Å to 3500 Å have been recently reported\textsuperscript{5} indicating that in this thickness range all the silver films exhibit the same behaviour. The $R_{HF}$ value is independent of the thickness $a$ and varies linearly with temperature\textsuperscript{5} (Figure 2).

It is not surprising that a constant value of $R_{HF}$ was obtained in silver films of thickness $a > 500$ Å; the F–S theory predicts that the “ordinary” size effect vanishes when $k$ approaches 1 (i.e. for $a > 400$ Å, $R_{HF} \approx R_{HO}$).

In comparing these results with the theory, we may deduce for $\beta_{RH}$ (as defined by Eq. 13) a value of

$$\beta_{RH} \approx +1.25 \cdot 10^{-3} \text{K}^{-1}$$

As the t.c.r. of an infinitely thick silver film is equal to $2.45 \cdot 10^{-3} \text{K}^{-1}$ \textsuperscript{9} it is clear from the above discussion (Section 2) that the contribution of the external surface scattering on the $\beta_{RH}$ coefficient becomes negligible in this thickness range. Thus we may verify a qualitative hypothesis previously advanced by the authors; indeed we have quantitatively demonstrated that for $k > 1$ the change in $R_{HF}$ with temperature is due to the variation of electronic density $n$.

It should be mentioned that, for metallic films for which quasi-free electron model is valid, no experimental works have been reported in recent years.

However as no disagreement has been observed between the present analysis and experimental studies, we estimate that the temperature dependence of $R_{HF}$ may be understood in terms of $n(T)$ which is generally a monotonic function of temperature for example\textsuperscript{5,4,8} but also in terms of $l_0(T)$ (i.e. $k(T)$) for thin films (i.e. $k < 1$ in the case of diffuse scattering) for which we have to consider the external surface scattering that comes into play.

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