Schottky law rather than the Poole-Frenkel law. It has been shown that any model invoked to explain the above anomaly must of necessity include not only trapping centers but also donor centers. We have proposed a model on this basis in which we have shallow neutral traps and deep donors. This model exhibits a bulk conductivity which is field-dependent in a manner usually associated with the Schottky effect even though the Poole-Frenkel effect is the operating mechanism, thus resolving the "anomalous" Poole-Frenkel effect in Ta_2O_5 and SiO films. It is shown that this is the only simple system of a single trap[11] and donor level that will exhibit the anomalous Poole-Frenkel effect.

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Size Effects in the Longitudinal Magnetoresistance of Thin Silver Films

K. L. Chopra

Ledgemont Laboratory, Kennecott Copper Corporation, Lexington, Massachusetts
and
National Magnet Laboratory, Massachusetts Institute of Technology, Cambridge, Massachusetts*

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Size effects in the longitudinal magnetoresistance of metal films have been observed at low temperatures and high magnetic fields. Partial specular reflection of conduction electrons is observed in silver films, provided the ratio α of the thickness to the effective-electron mean free path is smaller than 0.5. It is shown that the scattering coefficient (proportion of specularly reflected conduction electrons) can be determined from the size-effect data. Size effects for films with $\alpha>0.5$ are in agreement with total diffuse-scattering behavior. The dependence of the scattering coefficient on the value of α suggests that the nature of scattering depends on the angle of incidence of the electrons at the surface.

INTRODUCTION

THE scattering of conduction electrons from the surface of a metal film of thickness comparable in magnitude to the mean free path (l) has generally been considered[1] to be diffuse (inelastic). Recently, however, the studies of the thickness dependence of the electrical conductivity by Chopra and co-workers,[2–5] Larson and Boiko,[6] and Lucas[7] have established that the scattering in vacuum-evaporated gold and silver films is partially specular (elastic). An unambiguous determination of the fraction of specularly reflected electrons (the scattering coefficient p) from conductivity measurements on various films is difficult. The assumptions of a constant free path and a scattering coefficient for all the films is hardly justified, since each film has a characteristic value of l and also, presumably, of p. A meaningful scattering coefficient can be determined only for one film, either by changing l by varying the temperature[5] or by modifying the geometrical trajectories of the conduction electrons so as to change the amount of scattering. The latter corresponds to the galvanomagnetic size effects which are easily swamped by the large-bulk galvanomagnetic effects in noble metals. The bulk longitudinal magnetoresistance[8] generally increases to saturate to a small constant value at high magnetic fields. Deviations from this behavior caused by the geometrical size effects could, therefore, be observed and measured. The necessary condition to satisfy is: $l>t$ (film thickness)$>r$ (orbital radius of the electron trajectory under the applied magnetic field). This is now possible, due to the availability of high magnetic fields and epitaxially grown metal films of long mean free paths at low temperatures.[5,6]

Longitudinal magnetoresistance size effects are also expected to throw some light on the question as to what determines diffuse scattering. We know that although the polycrystalline and epitaxially grown continuous films have similar surface smoothness, the latter exhibit more specular reflection.[4,5] It is conceivable, as already suggested by Parrott,[9] that, in analogy with the reflection of light, specular reflection takes place only

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if the angle of incidence of the electron at the film surface is smaller than a critical value. This would imply that specular scattering is a function of the ratio of the mean free path to the film thickness.

We have observed size effects in the longitudinal magnetoresistance of thin films of gold, silver, indium, and aluminum at 4.2°K under applied fields up to 150 kG. The size effects reported here in this paper on silver films are typical of other films, although more marked in magnitude due to the longer mean face paths available.

EXPERIMENTAL DETAILS

Thin films were prepared by evaporation of 99.9999% pure silver from a tungsten basket in a vacuum of $\sim 10^{-6}$ Torr. Single-crystal films were grown epitaxially by depositing at a rate of ~ 10 Å/sec onto freshly cleaved mica substrates maintained at 150°C. Films deposited on cleaved mica and ultrasonically cleaned glass substrates at room temperature were polycrystalline. In some cases, films were annealed at 150°C after deposition. The film thickness was determined gravimetrically and checked interferometrically. Well-defined rectangular strips of films were mounted on a suitable holder which could be aligned accurately inside the Dewar filled with liquid helium. The resistivity was measured using the four-probe technique. The variable longitudinal magnetic field was produced by a 150-kG Bitter solenoid available at the National Magnet Laboratory. The magnetoresistance as a function of the magnetic field was recorded by an x-y recorder.

RESULTS

The data shown here have been chosen to illustrate the well-defined cases of total specular, partial specular, and total diffuse scattering of conduction electrons. The magnetic-field variation of the longitudinal magnetoresistance $\Delta\rho/\rho$ (ρ is the zero-field resistivity of the corresponding film at 4.2°K) for a 2.68-μ (111) epitaxially grown and a 1.35-μ annealed polycrystalline silver film are shown by circles and diamonds, respectively, in Fig. 1. Similarly, the data on a 1.2-μ thick unannealed silver film deposited on glass substrate are shown by circles in Fig. 2. The value of $\alpha = t/l$, which characterizes each film, has been determined by calculating the effective mean free path l from the Drude-Lorentz relation based on the free-electron theory:

$$\sigma_{4.2°\mathrm{K}} = (ne^2/m\bar{v})l_{\mathrm{eff}}, \tag{1}$$

where $m\bar{v}$ is the average momentum of conduction electrons at the Fermi surface, n is the number of conduction electrons per unit volume, and e is the electronic charge. The effective values of mean free path for 2.68-, 1.35-, and 1.2-μ thick films are about 53, 13, and 2.4 μ.

The abscissa $\beta = t/r$ is proportional to the strength of the applied magnetic field. The radius of curvature of

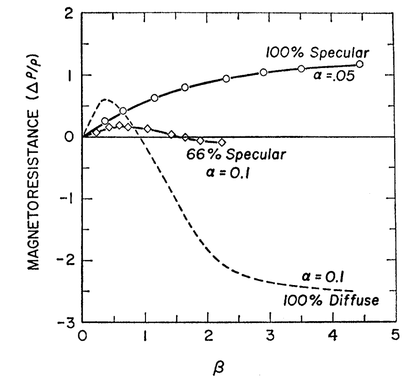

FIG. 1. Longitudinal magnetoresistance at 4.2°K as a function of $\beta = t/r$ for a 2.68-μ epitaxially grown Ag/mica (111) film (○ ○ ○); and for a 1.35-μ polycrystalline silver film (◇ ◇ ◇). The dashed curve is the theoretical curve for perfect diffuse scattering corresponding to $\alpha = 0.1$, and is derived from Kao's calculations.

the free electron r in a magnetic field H is given by

$$r = m\bar{v}c/eH \sim \frac{8\ \mathrm{Oe}}{H}\ \mathrm{cm}.$$

DISCUSSION AND INTERPRETATION

A generalized theory[10] of galvanomagnetic size effects for a general Fermi surface, anisotropic relaxation time, and an arbitrary scattering coefficient does not exist at present. The size effects, which are essentially geometrical and classical in nature, have been calculated on the assumption of a free-electron model and an isotropic Fermi surface, even though these assumptions are inadequate, because they lead to zero bulk magnetoresistance. We can, however, interpret our results by comparing the data with the observed bulk (specular) magnetoresistance and the theoretically calculated geometrical contribution to the magnetoresistance for the diffuse-scattering case.

The simplified calculations of the longitudinal-magnetoresistance size effect by Koenigsberg,[11] following Chamber's treatment for the case of wire geometry, yield no effect for $t/r \gg 1$. For $t/r \ll 1$,

$$\rho_{\mathrm{Film}} = \rho_{\mathrm{Bulk}}[1 + (3\pi/8\beta)]. \tag{2}$$

That is, at high fields ($\beta \gg 1$), the surface contribution vanishes as expected on the simple physical picture of the electron trajectories.

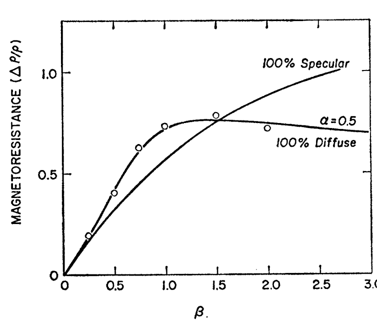

FIG. 2. Longitudinal magnetoresistance at 4.2°K as a function of β for a 1.2-μ polycrystalline silver film of $\alpha = 0.5$. The solid curve for 100% specular reflection corresponds to that of Fig. 1. The 100% diffuse scattering curve for $\alpha = 0.5$ is derived from Kao's calculations.

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Using similar formulation, but more detailed electron trajectories, Kao[12] has obtained a solution of the same problem by numerical integration. His calculations for total diffuse scattering predict an initial enhancement of the magnetoresistance followed by a sharp decline and an asymptotic approach to the bulk zero-field value. Theoretical curves for 100% diffuse scattering for $\alpha=0.1$ and $\alpha=0.5$ are shown in Figs. 1 and 2, respectively. These curves have been derived by adding the size-effect contribution given by Kao's theory for 100% diffuse scattering to the 100% specular or bulk behavior (indicated in both figures) observed for silver by Lüthi.[8]

A comparison of the experimental data with the theoretical curves shows that whereas data on 2.68-μ thick epitaxially grown silver film with $\alpha=0.05$ fit perfect specular behavior, that of unannealed 1.2-μ thick polycrystalline silver film (Fig. 2) with $\alpha=0.5$ agree well with the perfect diffuse-scattering case. The initial enhancement of magnetoresistance predicted by Kao's theory is observed in the latter case.

The behavior that is commonly observed for most annealed polycrystalline films is that shown by data on 1.35-μ thick silver film with $\alpha=0.1$. It is clearly a partial specular case. The observed variation, e.g., a rise to a maximum and then a decrease to a constant value, is expected in such a case. No enhancement of the magnetoresistance over the bulk value, however, is observed. The position of the peak in the magnetoresistance curve is found to depend on α rather than β. This is contrary to what one expects on purely geometrical considerations. Similar observations have also been made on bulk antimony[13] and bismuth.[14] Thus, the lower the value of α, the smaller is the value of β required to obtain maximum magnetoresistance. This clearly suggests that diffuseness of scattering of conduction electrons is a function of $\alpha=t/l$. Films with α smaller than 0.5 have been found to exhibit partial specular scattering. This indicates that partial specular reflection takes place if the angle of incidence of electrons at film surface ($\sim l/t$) is less than 30° for silver films. Although more quantitative data are necessary to yield reliable functional dependence, the merit of Parrott's suggestion is clearly indicated.

It is possible to determine the scattering coefficient p from the data on the partial specular case. The resistivity of the film is given[1] by

$$\begin{aligned}\rho_{\mathrm{Film}}&=\rho_{\mathrm{Bulk}}+\rho_{\mathrm{Surface}}+\rho_{\mathrm{Imp}}\\&=\rho_{\mathrm{Bulk}}\left(1+\frac{3}{8}\frac{1-p}{\alpha}\right)+\rho_{\mathrm{Imp}}\,,\end{aligned}\tag{3}$$

where ρ_{Imp} is the contribution due to impurities and imperfections. Since $\rho_{\mathrm{Bulk}}=\rho_{\mathrm{Specular}}$ and $\rho_{\mathrm{Bulk}}(1+3/8\alpha)=\rho_{\mathrm{Diffuse}}$, we can rearrange Eq. (3) as

$$\rho_{\mathrm{Film}}=p\rho_{\mathrm{Specular}}+(1-p)\rho_{\mathrm{Diffuse}}+\rho_{\mathrm{Imp}}\,.\tag{4}$$

The various scattering contributions are, therefore, additive. We expect the same to be true for the magnetoresistance contributions. The experimental curve can thus be reconstructed by adding the 100%-diffuse and 100%-specular curves, using an appropriate value of p. This method yields an average value of $p=0.7$ for $\beta>1$.

We can also calculate p as follows: From Eq. (3), the magnetic-field variation of resistivity is

$$\Delta\rho_{\mathrm{Film}}=\Delta\rho_{\mathrm{Bulk}}+\Delta\rho_{\mathrm{surface}}\,.\tag{5}$$

Here, we assume that $\Delta\rho_{\mathrm{Imp}}=0$.

At high fields ($\beta>1$), $\Delta\rho_{\mathrm{Surface}}\to\rho_{\mathrm{Surface}}$. Thus,

$$\begin{aligned}\rho_{\mathrm{Surface}}&=(\Delta\rho_{\mathrm{Film}}-\Delta\rho_{\mathrm{Bulk}})\\&=\rho_{\mathrm{Bulk}}\frac{3}{8}\frac{1-p}{\alpha}\,.\end{aligned}\tag{6}$$

The experimentally determined $\Delta\rho$ in Fig. 1 is small, and appears to have saturated at $\beta=2.25$, allowing further extrapolation without a significant error. The calculated value of p at high fields is 0.66 (66% specular) and is thus in close agreement with the value found by using Eq. (4).

CONCLUSIONS

It is concluded that the size effects in the magnetoresistance of thin films can be observed and interpreted to explain surface scattering of conduction electrons.

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