Applied Surface Science 332 (2015) 694–698

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Resistivity and Hall voltage in gold thin films deposited on mica at room temperature

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ARTICLE INFO

Article history:
Received 9 September 2014
Received in revised form
12 December 2014
Accepted 21 January 2015
Available online 29 January 2015

Keywords:
Resistivity
Hall voltage
Thin film
Gold

ABSTRACT

We report the thickness dependence of the resistivity measured at 4 K of gold films grown onto mica at room temperature (RT), for thickness ranging from 8 to 100 nm. This dependence was compared to the one obtained for a sample during its growth process at RT. Both behaviors are well represented by the Mayadas–Shatzkes theory. Using this model, we found comparable contributions of electron surface and electron grain boundary scattering to the resistivity at 4 K. Hall effect measurements were performed using a variable transverse magnetic field up to 4.5 T. Hall tangent and Hall resistance exhibit a linear dependence on the magnetic field. For this magnetic field range, the Hall mobility is always larger than the drift mobility. This result is explained through the presence of the above-mentioned scattering mechanisms acting on the galvanomagnetic coefficients. In addition, we report the temperature dependence of the resistivity between 4 and 70 K.

1. Introduction

Metallic films deposited on insulating substrates are one of the most used systems in the study of optical, magnetic, mechanical and electrical properties of metals [1]. Control over sample thickness and microstructure, through modification of growth conditions, increases the usefulness of these systems. Among them, gold thin films on an insulating substrate are one of the most used due to easy fabrication and to the null oxidation at atmospheric conditions [2].

From the point of view of electrical transport, in gold thin films at room temperature, a series of scattering mechanisms participates increasing the resistivity: electron phonons scattering, electron grain boundary scattering and electron surface scattering. Some reports show experimentally the effect of each mechanism. Some examples, Kastle et al. [3] and Henriquez et al. [4] reported gold thin films where the dominant mechanism is electron surface scattering. They found that the temperature dependence of the resistivity exhibits an increase of its slope respect to that of the bulk. On the other hand, when the dominant mechanism is electron grain boundary scattering this increase is not observed, and also, the mean free path at 4 K correspond to mean grain diameter [5].

Deposition of thin gold films at room temperature results in samples characterized by a mean grain diameter comparable to the film thickness. Also, this dimension increases as thickness grows. Some examples of these morphological conditions: thin gold films on mica [2], on SiO_2 [6] and on glass [7]. Thereby, these samples present an opportunity to study the effect of both mechanisms, electron surface scattering and electron grain boundary scattering, acting on the resistivity and on the galvanomagnetic coefficients.

In this work, we studied the resistivity, magnetoresistance and Hall voltage and its relation to the microstructure, in a set of gold thin films where the electron grain boundary scattering and electron surface scattering affect the charge transport process.

2. Experimental

We deposited thin gold films onto freshly cleaved mica through thermal evaporation at room temperature. Starting from 99.9999% pure gold, we evaporated at 1.2 nm/min from a tungsten basket in a High Vacuum chamber (10^{-4} Pa). A quartz microbalance located close to the sample monitored the evaporation rate. Mica surface roughness is about 0.3 nm over areas of 1 μm × 1 μm. Sample thickness was measured by Tolansky Optical Interferometry performed over test samples on glass slides placed close to the mica substrate.

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http://dx.doi.org/10.1016/j.apsusc.2015.01.166
0169-4332/

Thicknesses are consistent with values measured by the quartz microbalance.

Morphology of the gold films was characterized through scanning tunneling microscopy (STM) at room temperature (RT) in air using Pt-Ir tips. Images were processed with linear plane fit in order to remove the tilt with WSxM [8] before the statistical analysis. Using ImageJ software we marked grain boundaries and measured the enclosing area. We measured over 500 grains per sample to ensure a representative value [9]. Then, the characteristic size of a grain is the diameter of a circle enclosing the mean area.

Regarding to electrical characterization, in all samples we used the 4-contact method. We performed three different measurements:

(a) Determination of the temperature dependence of the resistivity between 4 and 70 K. Samples were located in a copper block inserted in a superconducting Magnet built by Janis Research Co. The electrical measurement started when the standard deviation of the temperature was smaller than 0.1 K. Films were fed with alternate current of 210 Hz and smaller than 500 μA. Voltages signal were acquired using computer controlled 830's LIA built by Stanford Research.
(b) Determination of magnetoresistance and Hall Voltage at 4.2 K. Experimental set up is similar to (a), but samples were immersed in magnetic fields up to 4.5 T. To measure the Hall voltage, we used the five-contact method, seeking to null the transverse signal in the absence of a magnetic field. Details of the method can be found elsewhere [10]
(c) Determination of the resistance during the evaporation process at RT. We pre-evaporated gold contacts on mica, and measured the resistance during the film evaporation. The sample was fed with direct current, alternating the polarity each 4 s approximately. Voltages were acquired through Nanovoltmeters built by Keithley.

3. Theory

3.1. Resistivity

In 1970, Mayadas and Shatzkes (MS) published the first theory of resistivity including the effect of both electron-grain boundary and electron-surface scattering [11]. The theory describes the electron motion using a Boltzmann Transport Equation. Grain boundaries are represented by a series of Dirac δ function potentials and characterized by a reflectivity coefficient R. This coefficient represents the fraction of electron reflected specularly at the grain boundary. The distance separating the Dirac δ function potentials is distributed following a Gaussian, characterized by an average separation d and a standard deviation s. The effect of electron-surface scattering on the resistivity is introduced through a specularity coefficient P based on Fuchs-Sondheimer theory [12]. This coefficient is imposed on the electron distribution function as a boundary condition at the interfaces that limit the metallic film. $(1-P)$ represents the fraction of the charge carrier scattered by the surface, which induce an increase on the resistivity. We modified this boundary condition allowing two different specularity parameters P and Q, associated to the upper and lower interface, respectively, following the work published by Lucas [13]. Finally, the expression to calculate the resistivity increase ρ_F/ρ_0 (where ρ_F represents the film resistivity and ρ_0, the bulk resistivity), depend on seven parameters: d, s, R, P, Q, t and ℓ_0. The last two symbols represent the thickness and the bulk mean free path, respectively. The final expression used for calculating the resistivity was published elsewhere [4,5]

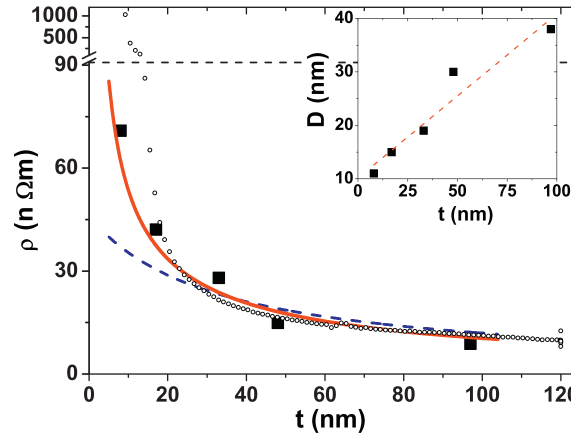

Fig. 1. Thickness dependence of the resistivity at 4 K. Squares represent the experimental data. Empty circles represent the resistivity determined by the in situ measurement of the resistance during the evaporation process at room temperature. An offset was subtracted to get the resistivity at 4 K in the same sample. Continuous and dashed lines represent the best fit obtained by Mayadas–Shatzkes–Lucas theory (MSL), for $(R, P, Q) = (0.18, 0.0, 0.0)$ and $(0.24, 0.0, 1.0)$, respectively. Inset: Thickness dependence of the mean grain diameter for the samples labeled with squares. Dashed line represents the linear dependence used in MSL.

3.2. Magnetoresistance and Hall voltage

In this work, we study the transverse magnetoresistance $\Delta\rho/\rho = [\rho(B) - \rho(B \equiv 0)]/\rho(B \equiv 0)]$, where $\rho(B)$ represents the resistivity when the sample is immersed in a transverse magnetic field B. To our knowledge, there is not a theory including electron-grain boundary scattering, electron-surface scattering and magnetic field describing the magnetoresistance in thin metallic films. Then, the classical interpretation of the effect of the magnetic field on the conduction electrons will be considered. The trajectories between scattering events are modified due to the curvature induced by the magnetic field. A measurement of this is the fraction of cyclotron orbit performed by the conduction electrons $\omega\tau$, where τ is the scattering time and $\omega = qB/m$ (q and m are the charge and mass of conduction electrons, respectively, and B, the magnetic field). If $(\omega\tau) \ll 1$, then the effect should be weak. In this case, magnetoresistance is proportional to $(\omega\tau)^2$, i.e., there is a quadratic dependence on the magnetic field.

Regarding the Hall voltage, the drift velocity v_D of the electrons in presence of the electric field $\mathbf{E}$, can be expressed as $\mathbf{E}$, where $v_D = \mu_D\mathbf{E}$, where μ_D represents the drift mobility. To add a magnetic field $\mathbf{B}$, the Lorentz force on the conduction electrons modify the drift velocity such that $v_D = \mu_D\mathbf{E} + \mu_D\mu_H\mathbf{E} \times \mathbf{B}$, where μ_H represents the Hall mobility. In the particular case where $\mathbf{E} = (E_X, E_Y, 0)$ and $\mathbf{B} = (0, 0, B)$, canceling the component v_{DY} of the drift velocity leads to the Hall tangent $tg(\theta) = E_{Hall}/E_{Longitual} = \mu_H B$ On the other hand, if we only want to compare the Hall voltage from different samples, the Hall resistance $R_H = V_H/I$ is often used, where I is the current feeding the sample.

4. Results

A resume of the morphological and electrical characterizations of gold thin films appears in Table 1.

Fig. 1 shows the thickness dependence of the resistivity at 4 K. Using Mayadas–Shatzkes–Lucas theory (MSL), we adjusted experimental data following procedure outlined below. First, we adjusted the thickness dependence of the mean grain diameter $d(t)$ from experimental data determined by STM. Inset of Fig. 1 shows the thickness dependence of the mean grain diameter and the linear regression: $d(t) = 0.31t + 10.14$ (d and t in nanometer).