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A method to investigate the electron scattering characteristics of ultrathin metallic films by *in situ* electrical resistance measurements

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In this article, a method to measure the electrical resistivity/conductivity of metallic thin films during layer growth on specific underlayers is described. The *in situ* monitoring of an underlayer electrical resistance, its change upon the incoming of new material atoms/molecules, and the growth of a new layer are presented. The method is easy to implement and allows obtaining *in situ* experimental curves of electrical resistivity dependence upon film thickness with a subatomic resolution, providing insight in film growth microstructure characteristics, specular/diffuse electron scattering surfaces, and optimum film thicknesses. © 2009 American Institute of Physics.
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I. INTRODUCTION

In situ electrical resistance measurements have been used to investigate the surface chemistry and growth of heterostructures prepared by atomic layer deposition¹ to optimize the giant magnetoresistance ratio of spin valves²⁻⁴ and to investigate and optimize the formation (growth of Al and its oxidation) of Al₂O₃ magnetic tunnel junctions prepared by evaporation⁵ and by magnetron sputtering.⁶ Current trends in magnetoelectronic and spintronic devices indicate the need of heterostructures composed of nanolayers with increasing reduction of layer thickness, where size effects play an important role. Continuous *in situ* electrical resistance measurements is a suitable and expedite way to understand the growth mechanisms and to optimize the electron scattering characteristics of such nanostructured films.

II. EXPERIMENTAL METHOD

The thin films investigated in this work by the present *R in situ* method were prepared by ion beam deposition, in a Commonwealth Scientific 1160L system. The unit consists of a high vacuum deposition chamber with a base pressure of 10⁻⁷ Torr, pumped down by a cryopump. The deposition chamber uses a hot filament Kauffman source with a 3 cm Mo grid, which focuses the beam into a target. The target stage consists of a cube, integrating four different targets, having each a diameter of 75 mm. A substrate table, consisting of a rotating arm with a base plate at its extremity, places the substrate near the target of material to be deposited. The normal to the substrate plane and the normal to the target plane make an angle between 180° (substrate facing the target) to 225° (substrate horizontal). A gas flow controller inserts an inert gas into the ion gun, where the cathode filament starts a plasma with a discharge current, I_D . The beam is then extracted by applying voltages to the anode and grid, V_B = beam voltage and V_A = accelerator voltage, respectively. The films were prepared with the following deposition pa-

rameters: I_D = 2.2 A, V_B = 850–1000 V, V_A = 100 V, and chamber argon pressure 2.3×10^{-4} Torr. All films use Corning glass substrates and a basecoat/underlayer of amorphous Ta. The targets, the chamber walls, and substrate base plate are water cooled with chilled water at a temperature of about 14 °C.

The *in situ* setup uses electrical resistance measurements in the Van der Pauw geometry,⁷ similar to that described by Schuisky *et al.*¹

For the *R in situ* experiments, four electrical contacts need to be first defined on the substrate where the film will be deposited. These consist each of a circular pad and a narrow stripe extending toward the center of the substrate, as shown in Fig. 1. The narrow stripes (fingers) allow electrical contact between the pads and the film to be investigated. The electrical contacts are defined with a stencil aluminum mask (shadowing mask) and consist of Ta 100 Å/Al 400 Å/Ru 30 Å. Once the substrates with electrical contacts are defined, an electrical probe cap made of a ceramic material (Macor), integrating four contact probes and having a circular hole in the center, is loaded over the substrate. The contact probes are aligned with the center of the pads of the substrate, and the cap hole is centered with the four metallic stripes in a manner that the basecoat of Ta, underlayer, and magnetic layers form an homogeneous circular disk, with a diameter of approximately 10 mm, as shown in Fig. 1. The electrical stripes, extending from the pads, will make electrical contact to the film in four edges, in a nearly symmetrical manner. Cross-section scanning electron microscopy (SEM) observations in the regions substrate/magnetic film and substrate/contacts/magnetic film are shown in Fig. 1. The micrographs in Fig. 1 were obtained in the backscattering electron detection (BSED) mode, where materials with higher Z number give a brighter contrast in the images, while the lighter elements give a darker contrast. The SEM analysis indicates that the Ta 100 Å/Al 400 Å/Ru 30 Å electrical contact stripes have extended transition regions (as opposed to 90° step film

FIG. 1. (a) Photographs of the glass substrates used in the experiments: (left) with electrical contacts of Ta 100 Å/Al 400 Å/Ru 30 Å, predefined in the substrates before the experiments, (right) with electrical contacts and circular shape Ta basecoat/underlayer/film, after the experiment was concluded. (b) SEMs, in BSED mode, in the cross-section geometry: (left) region glass/Ta 100 Å/Al 400 Å/Ru 30 Å/Ta 70 Å basecoat/underlayer/magnetic film, (right) region glass/Ta 70 Å basecoat/underlayer/magnetic film.

profile) to the glass substrate, with a length of about 30 μm , much larger than the thickness of the films involved, but much smaller than the diameter (10 mm) of the sample electrically monitored.

Electrical wires connect the contact probes to an electrical feedthrough with four wires connecting to a Keithley sourcemeter. The substrate and electrical probe cap are clamped to the substrate table of the deposition chamber. The substrate table has a manual mechanical shutter, electrically actuated by a switch and compressed air, which takes about 1–2 s to completely open or close. The shutter controls the beginning and ending of each deposition. The first material deposited is the basecoat of Ta. Initially, the shutter is open during an interval of time to have deposited on the substrate a layer of Ta 20 Å, which provides electrical connection between the four electrical contact stripes. The sourcemeter is then turned on, in the four-wire Ohmmeter mode, using two contiguous pads to apply current and the remaining two to measure voltage. The typical resistance value of Ta 20 Å is 1 k Ω . A LABVIEW program is then started to perform the R *in situ* data acquisition, consisting of the Ohmmeter reading as a function of time, for any two intervals of time. The minimum sampling time is 600 ms, but 1 s sampling time was used. During an interval of time of about 10 s, the electrical resistance of the sample is monitored with the shutter closed. Then, the shutter is open and the time of deposition starts counting (also monitored by a separate chronometer) until the time to complete the layer is reached and the shutter is closed. The absence of a synchronization mechanism between the opening of the shutter event and the clock of the resistance monitoring data acquisition produces an error, $\varepsilon_{\text{synch}}$, addressed next. The program continues running a little longer time, until it stops the data acquisition, and waits for the user to acknowledge that the monitoring of the electrical

TABLE I. Deposition rates in Å/s.

	Ta	Ru	Co ₈₅ Fe ₁₅	Ag	Co ₆₀ Fe ₂₅ B ₁₅
Dep. rate (DR)	0.56/0.3	0.32	0.8	0.55	0.75

resistance for the next layer deposition can be started. During this period of time, the user can turn off the beam, choose another target material to be deposited, restart the beam, and adjust the beam parameters. The intervals of measurement of resistance, corresponding to the state of the shutter closed allows one to estimate the resistance measurement error, given by $|2\sigma|$, where σ is the standard deviation.

The resistivity of each layer as a function of the layer thickness, l , is obtained from the experimental data, with the following expression, corresponding to the parallel resistance configuration:

$$\rho_{\text{layer}}(l) = \text{GF} \frac{R_0^{4w} \times R^{4w}(t)}{R_0^{4w} - R^{4w}(t)} \times t \times \text{DR}, \quad (1)$$

where R_0^{4w} is the electrical resistance of the sample (corresponding to the buffer/underlayer) before the shutter is open (before the layer deposition starts), $R^{4w}(t)$ is the resistance value at the deposition instant t , DR is the calibrated deposition rate, $l = t \times \text{DR}$ is the layer thickness, and GF is the geometric factor.

Assuming that the film growth is homogeneous in the area of measurement so that GF is constant, the errors in the experimental curves of ρ_{layer} versus layer thickness, l can be estimated by $\varepsilon = 4 \times \varepsilon_R + \varepsilon_{\text{synch}}$. The error in the measurement of R , $\varepsilon_R \cong 2\sigma/\langle R \rangle < 0.2\%$ is negligible in relation to the error in the deposition time absolute value, $\varepsilon_{\text{synch}}$. In the worst case scenario, the shutter opens when the resistance monitoring just finished one cycle of 1 s period, leading to a major error in time counting $\varepsilon_{\text{synch}} \cong 0.6$ s, involving each measurement at least 300 ms integration time. In this case, a very large absolute error is present in the resistivity values obtained in the first seconds of deposition, but decreases as $\varepsilon_{\text{synch}}/t$ with the deposition time, t , increases.

In general, the error is less than 5% after 10 s deposition time. Nevertheless, when this error is large, it leads to a discrepancy in the trend of the curve. In this case the first data points of resistance acquired after the shutter is open are rejected.

The deposition rate is obtained by measurements of film thickness divided by its deposition time. The calibration films have thicknesses higher than 2000 Å, to minimize to less than 10% the error in the thickness measurement obtained by profilometry. In Table I are listed the deposition rates for the materials used.

III. EXPERIMENTAL RESULTS AND DISCUSSION

The raw experimental data corresponding to *in situ* electrical resistance variation with time are presented in Fig. 2. When the data acquisition starts, the beam is on and the shutter is closed, and the electrical resistance corresponding to Ta 20 Å is measured and is nearly constant, with a value $R^{4w} \cong 1$ k Ω . Next, the shutter is open, and atoms of Ta are

FIG. 2. Raw experimental data of *in situ* electrical resistance as a function of time, corresponding to the deposition of Ta 50 Å/Ag 200 Å on a glass substrate.

deposited over the Ta 20 Å underlayer until the Ta basecoat reaches a thickness $l_{\text{Ta}} = 70$ Å and the shutter is closed. The electrical resistance is then $R^{4w} \approx 100 \Omega$ and it drops to $R^{4w} \approx 1 \Omega$, after a layer of Ag with a thickness of 200 Å is deposited. By assuming that the geometrical factor in Eq. (1) is constant and has a value of approximately 3, the following values of resistivity are obtained: $\rho(\text{Ta } 70 \text{ Å}) = 240 \mu\Omega \text{ cm}$ and $\rho(\text{Ag } 200 \text{ Å}) \approx 7 \mu\Omega \text{ cm}$. The resistivity value of the Ta film is close to that obtained in thick films prepared in similar conditions, although amorphous Ta films with resistivity values of $200 \mu\Omega \text{ cm}$ (Ref. 8) and $145 \mu\Omega \text{ cm}$ (Ref. 9) have been reported. The electrical resistivity of a polycrystalline thin film of Ag is expected to have a much higher value than the bulk ($\rho_{\text{Ag,bulk}} = 1.6 \mu\Omega \text{ cm}$),¹⁰ mainly due to size effects¹¹ and grain size electron scattering.^{12,13} Nevertheless, the electrical resistivity of Ag 200 Å obtained is about twice than that of films prepared by magnetron sputtering, with columnar structure and high (111) texture.¹⁴

The repeatability and sensitivity to the chamber base pressure of the *in situ* electrical resistance experimental data were addressed by depositing in identical beam conditions three samples of Ta 70 Å/Ag 200 Å. The experimental results presented in Fig. 3 show good repeatability, but indicate that the chamber vacuum pressure significantly affects the silver scattering characteristics, for $l_{\text{Ag}} \leq 120$ Å. *Ex situ* resistivity data, obtained with narrow stripes and four electrical contacts, two inner contacts to measure voltage and two outer contacts to apply uniform electrical current, are also shown [solid symbols, Fig. 3(b)]. The distance between the outer contacts and adjacent inner contacts is larger than three times the width of the stripe, to assure uniform electrical current distribution in the region where the electrical resistance is measured. Although the total resistance of the Ta basecoat does not show much difference, it is likely that the surface roughness of the basecoat may be considerably different when the vacuum pressure is poorer. The lower resistivity value obtained by *ex situ* measurements in comparison to those obtained by *in situ* electrical resistance measurements, obtained with samples integrating thin Ag layer as glass/Ta

FIG. 3. Experimental data of variation of *in situ* electrical resistance corresponding to three runs of sequential deposition of glass/Ta 70 Å/Ag 200 Å having annotated the vacuum pressure; (a) variation of raw resistance vs deposition time for the Ta basecoat; (b) variation of resistivity vs layer thickness for Ag, where solid square symbols correspond to *ex situ* measurements. Inset, detail of $\rho(\text{Ag})$ for $l_{\text{Ag}} > 100$ Å.

70 Å/Ag 50 Å also indicates that the surface scattering plays an important role in the initial part of the experimental resistivity versus thickness data obtained for the silver films.

The experimental curves of *in situ* electrical resistance variation with sequential deposition time (the time intervals corresponding to the shutter closed condition have been subtracted) for thin films of Ta 70 Å/Ru_{#1} 13 Å/Co₆₀Fe₂₅B₁₅#1 130 Å/Ru_{#2} 13 Å/Co₆₀Fe₂₅B₁₅#2 130 Å/Ru_{#3} 13 Å and of Ta 70 Å/Ru_{#1} 13 Å/Co₈₅Fe₁₅#1 200 Å/Ru_{#2} 13 Å/Co₈₅Fe₁₅#2 200 Å are shown in Fig. 4. At first sight, the two curves show similar trends, characterized by a sharp decrease in electrical resistance with film thickness increase (neglecting the thin Ru layers). Nevertheless, when the electrical resistivity of each layer is extracted with Eq. (1), one finds very important differences between the thickness dependence of the electrical resistivity of the Co₆₀Fe₂₅B₁₅ and the Co₈₅Fe₁₅ layers, as shown in Fig. 5.

The CoFeB layers exhibit similar trends in layer thickness growth, characterized by the following features: an initial sharp decrease in electrical resistivity, from values of approximately $150 \mu\Omega \text{ cm}$ to minimum values of $60 \mu\Omega \text{ cm}$ at $l_{\text{CoFeB}} \approx 40$ Å for CoFeB_{#1} (grown on Ta 70 Å/Ru 13 Å) and $l_{\text{CoFeB}} \approx 20$ Å for CoFeB_{#2} (grown on Ta 70 Å/Ru 13 Å/CoFeB 135 Å/Ru 13 Å); a first linear regime of resistivity increase with layer thickness at a rate of $0.4 \mu\Omega \text{ cm/Å}$ and $0.6 \mu\Omega \text{ cm/Å}$, for CoFeB_{#1} and CoFeB_{#2} layers, respectively; a second linear regime of resistivity increase with layer thickness at a rate of approximately $0.2 \mu\Omega \text{ cm/Å}$. The experimental curves of electrical resistivity versus layer thickness become parallel, for $l \geq 75$ Å, but CoFeB_{#2} exhibits higher resistivity, approaching that of the bulk. Effectively, the *ex situ* resistivity of a

FIG. 4. Raw experimental data of variation of *in situ* electrical resistance with sequential deposition time; (a) film composition is Ta 70 Å / Ru_{#1} 13 Å / Co₆₀Fe₂₅B₁₅#₁ 130 Å / Ru_{#2} 13 Å / Co₆₀Fe₂₅B₁₅#₂ 130 Å / Ru_{#3} 13 Å; (b) film composition is Ta 70 Å / Ru_{#1} 13 Å / Co₈₅Fe₁₅#₁ 200 Å / Ru_{#2} 13 Å / Co₈₅Fe₁₅#₂ 200 Å.

Co₆₀Fe₂₅B₁₅ film with a thickness of 2000 Å is 114 $\mu\Omega$ cm. The initial decrease in resistivity suggests that the boron atoms are going into defect sites of the underlayer, and CoFeB_{#1} mostly consists of small grains of polycrystalline CoFe alloy; hence, the impurity scattering caused by boron atoms is less than that in the bulk material. When the boron atoms cannot go into the defect sites of the underlayer and

into the boundaries of the new grains that are being nucleated and grown, they start going into the matrix of the grains. Consequently, in the matrix, the distance between the boron atoms become smaller, reducing the mean free path of the electrons and consequently the resistivity increases. CoFeB_{#2} may have available less defect sites to be occupied by the boron atoms, consequently exhibiting a resistivity increase at earlier layer thickness and achieving a resistivity value close to the bulk material at lower layer thickness.

The experimental curves corresponding to the thickness dependence of the electrical resistivity of the polycrystalline Co₈₅Fe₁₅ layers exhibit very different trends to those observed with the Co₆₀Fe₂₅B₁₅ layers, as shown in Fig. 5. In order to have a better understanding of the stability of the experimental data and sensitivity of the technique in the same figure are shown the raw data for each of the CoFe layers, prior to and in the beginning of the layer deposition, where negative time corresponds to the shutter closed condition. The resistivity versus thickness data, extracted with Eq. (1), shows in particular that for layer thicknesses larger than the bulk mean free path of the electrons, which in these alloys at ambient temperature is of the order of 50 Å,^{9,13} the resistivity of the layers decreases with layer thickness increase. For layer thicknesses, $l \leq 50$ Å, size effects¹¹ such as degree of specular/diffuse scattering at the bottom and top surfaces of the film may significantly affect the electrical resistivity of the films. Furthermore, the grain boundary scattering¹² is expected to be dominant. These two aspects are analyzed in another article.¹³ For $l < 50$ Å, the experimental curves indicate that CoFe_{#1} (grown on Ta 70 Å / Ru 13 Å) has considerably higher electrical resistivity than CoFe_{#2} (grown on Ta 70 Å / Ru 13 Å / CoFe 200 Å / Ru 13 Å), suggesting that the former follows a process of small grain nucleation and growth while the latter accommodates to the existing grains of the underlayer.

IV. CONCLUSIONS

The presented method is easy to implement and is sensitive to resistivity changes produced by less than a monolayer of material being deposited on specific underlayers. It allows obtaining the electrical resistivity dependence upon film thickness with a subatomic resolution, providing insight in the scattering characteristics of very thin films, connected to film growth mechanisms and specular/diffuse degree of their surfaces/interfaces. The method can be further improved by synchronization of the data acquisition time with the mechanical shutter that initiates the film deposition in the substrate/underlayer.

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FIG. 5. Experimental curves of variation of resistivity with layer thickness, calculated with Eq. (1); (a) for Co₆₀Fe₂₅B₁₅ layers of the film glass/Ta 70 Å / Ru 13 Å / CoFeB 130 Å / Ru 13 Å / CoFeB 130 Å / Ru 13 Å; (b) for Co₈₅Fe₁₅ layers of the film glass/Ta 70 Å / Ru 13 Å / CoFe 200 Å / Ru 13 Å / CoFe 200 Å. Detail of raw resistance vs deposition time in the range of 1–20 s, where negative time correspond to the condition of shutter closed, for layers; (c) CoFe_{#1} and (d) CoFe_{#2}.

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