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Study of the different magnetoresistance sources in Ag/Co multilayers

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Abstract

We report results on magnetoresistance and magnetic properties of sputtered Ag/Co multilayers and their relation to structural properties. We found two components of the magnetoresistance: isotropic and anisotropic. The first one is found to be related to cobalt particles at the interfaces between magnetic and nonmagnetic layers and also to cobalt particles diluted into the silver layers. The other contribution is related to ferromagnetic multidomain Co layers. The results on magnetoresistance and magnetization at low fields, and conductivity measurements, give clear proof of a transition from granular to continuous structure of the magnetic layer. For example, in a Ag/Co multilayer series with silver thickness of 20 Å, such a transition occurs for a cobalt thickness around 5 Å.

1. Introduction

Since the discovery of giant magnetoresistance (GMR) in Cr/Fe multilayers [1], many other multilayer systems have been investigated in depth, not only because of their potential technical applications, but also to gain an understanding of the fundamental mechanism of the magnetotransport properties [2, 3]. However, GMR is not restricted to multilayer systems and many granular alloy systems have also been the subject of in-depth study [4–6]. Deposition by a sputtering method enables one to grow alternating very thin magnetic layers and thicker nonmagnetic layers. For very small Co thickness, the magnetic layer breaks into a discontinuous layer. This is called a granular multilayer, which is the structure at the border between multilayers and granular alloys. GMR in granular multilayers is isotropic, i.e., it does not depend on the relative orientation of the electric current and applied magnetic field, shows insignificant hysteresis, and displays a GMR effect less than that in multilayer samples under the same magnetic field. The response to the magnetic field strongly depends on the

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microstructure of the layers. For this reason, magnetoresistance is used to correlate it with the microstructure of the multilayer [7–9]. On the other hand, GMR is very sensitive to some preparation parameters: magnetic and nonmagnetic layer thickness, interlayer roughness, grain size, etc [10, 11]. It is believed that the mechanisms responsible for GMR are based on spin-dependent electron scattering at the interlayers as well as within the magnetic layers [3, 12].

In this paper, we focus our attention on the relationship between magnetoresistive properties and the microstructure of the magnetic layer in Ag/Co multilayers, deposited by sputtering techniques, as both layer thicknesses are increased. Extreme care was taken to keep the growing parameters constant. This system, which could be at the border between multilayer and granular alloys, is highly immiscible at equilibrium, as can be inferred from the Ag/Co binary phase diagram. In principle, that ensures well-defined interfaces. We use magnetic, magnetotransport, and resistivity measurements to obtain information about the critical thickness associated with the transition from a granular structure to a continuous layer. This was analysed as a function of the magnetic layer thickness. The influence of the nonmagnetic layer thickness was also studied.

2. Experiment

The samples used in this study were prepared in a multitarget RF sputtering system equipped with Co (99.99%) and Ag (99.99%) targets. All samples were deposited on glass substrates at room temperature (RT) under an Ar partial pressure of 3×10^{-3} mbar. The background vacuum pressure was better than 5×10^{-7} mbar. The deposition rates were about 1.4 Å s^{-1} for both Co and Ag. The layer thickness growth was timed with computer-controlled shutters. Deposition started with a Co buffer layer and was completed after the corresponding Co layer of the 15th Ag/Co bilayer, $\mathrm{Co}(t_{\mathrm{Co}})[\mathrm{Ag}(t_{\mathrm{Ag}})\text{–}\mathrm{Co}(t_{\mathrm{Co}})]_{15}$ had been produced, where t_{Co} and t_{Ag} are the cobalt and silver thicknesses, respectively. We have grown different series of multilayers with fixed Ag thicknesses of 10, 20, 30, and 40 Å, where t_{Co} ranges from 2 up to 20 Å. Additionally, series with $t_{\mathrm{Co}} = 10$ Å and t_{Ag} from 2 up to 120 Å were deposited. Magnetization was measured using a vibrating sample magnetometer (VMS) with applied fields up to 15 kOe parallel to the sample plane. Magnetoresistance (MR) measurements were made in the plane of the films using the standard dc four-point method, with applied magnetic fields up to 15 kOe perpendicular and parallel with respect to the electrical current. The four puncture contacts are mechanically aligned with an accurate precision, which permits us to arrange the current path parallel or perpendicular to the applied magnetic field, by rotating the sample holder by 90°. The MR was defined as $\mathrm{MR}\% = [(\rho(H) - \rho(H = 0)) \times 100\%]/\rho(H = 0)$. The low-angle x-ray reflectivity (LAXR) was used to test the grade of rugosity and structure in the multilayers. The reflectivity scans were fitted with the REFS MERCURY refinement program to estimate the layer rugosity and check the layer thicknesses [13, 14]. Electrical resistivity was measured in some of the series from 8 to 300 K in a closed-cycle He cryostat.

3. Results and discussion

Two different contributions were detected in the magnetoresistance of the Ag/Co multilayers upon comparing the longitudinal and transverse (electric current parallel (LMR) and perpendicular (TMR) to the applied magnetic field H, respectively) magnetoresistance loops, measured using moderate fields. In figure 1, the field dependences of the MR of some samples from the series $[\mathrm{Ag}(10)\text{–}\mathrm{Co}(t_{\mathrm{Co}})]_{15}$ up to $H_{max} = 500$ Oe are given. As can be seen, for Co thickness lower than 4 Å, the LMR and the TMR are very similar, i.e., the MR does not depend

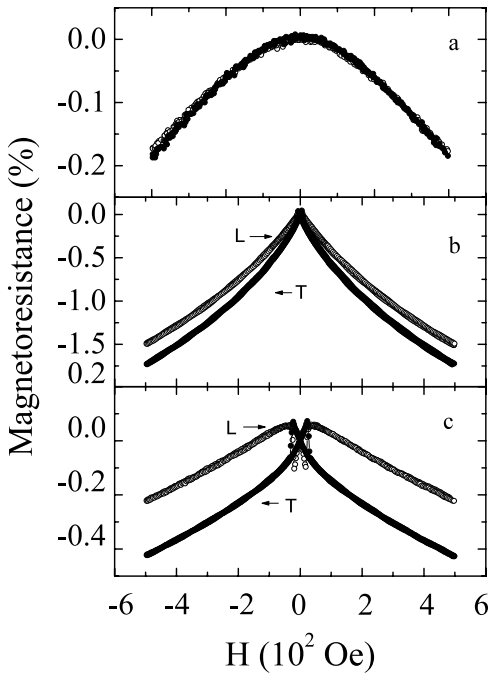

Figure 1. Magnetoresistance measurements for three Ag/Co multilayers from the series [Ag(10)–Co(t_{Co})]$_{15}$ where t_{Co} is (a) 3 Å, (b) 4 Å, and (c) 5 Å. L and T refer to longitudinal and transverse magnetoresistance in accordance with the usual definitions.

on the relative orientation of the current and H. That evidences the isotropic character of the MR in this range of thicknesses. Both measurements show negative magnetoresistance and broad MR%–H profiles without hysteresis. This behaviour of MR is usually found in granular systems [4, 15]. For Co thickness around 5 Å and Ag spacer thickness of 10 Å, a sudden change takes place in the MR measured at low fields (<75 Oe): LMR and TMR components display very dissimilar behaviours (see figure 1(c)). The shape of the MR–H curve shows positive LMR and negative TMR, displaying hysteresis at fields below 100 Oe (see figure 2). In this case, the MR depends on the relative orientation of the current and H, showing the anisotropic character at low fields when the Co layer thickness is above around 5 Å. Thus, LMR measurements show a change from negative to positive MR with increase of the magnetic layer thickness. A similar behaviour was also observed in other kinds of film [16]. The threshold value for that transition is between 4 and 7 Å of Co, depending on the series studied. Figure 2 shows magnetoresistance curves for two multilayers with $t_{Ag} = 6$ and 10 Å, and $t_{Co} = 10$ Å, with applied magnetic field up to 200 Oe. The appearance of that phenomenon in the LMR is remarkably sharp, allowing us to define a critical thickness t^*_{Co} which marks the transition from a granular to a quasicontinuous magnetic layer. On the other hand, the observed evolution of the TMR loop shapes (broad to a sharp drop at low field) as Co thickness increases is also in agreement with the reported transition from a granular to a continuous magnetic layer [17].

The critical thickness t^*_{Co} was also found to mark a noticeable change of regime in the magnetization loops $M(H)$ of the multilayers. Figure 3 shows the field dependence of the normalized magnetization for [Ag(40 Å)–Co(t_{Co} Å)]$_{15}$ multilayers with Co thicknesses of 5 and 60 Å, and [Ag(10 Å)–Co(t_{Co} Å)]$_{15}$ with Co thicknesses of 4 and 10 Å. Below t^*_{Co}, the loops show no hysteresis at all (figures 3(a) and (c)) and are far from saturation at 1 kOe. This

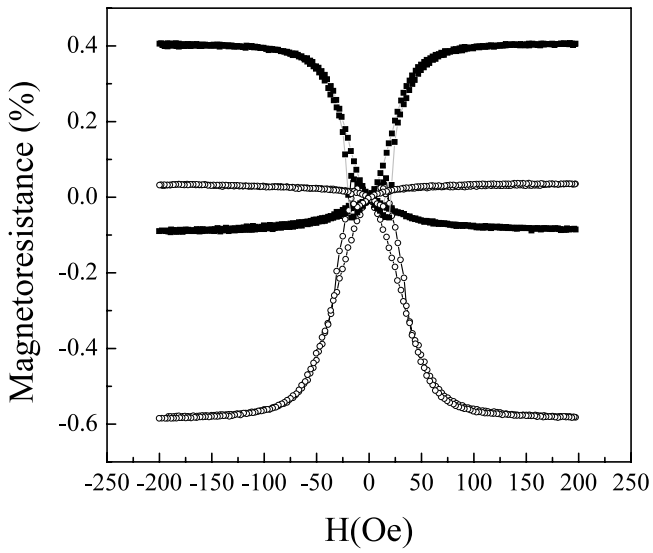

Figure 2. Magnetoresistance curves (MR) versus applied magnetic field (low field) for two multilayers: [Ag(6 Å)–Co(10 Å)]$_{15}$ (circles); and [Ag(10 Å)–Co(10 Å)]$_{15}$ (squares). TMR and LMR relate to the upper and lower curves, respectively.

is a characteristic superparamagnetic behaviour which points to the presence of Co particles responsible for the isotropic magnetoresistance [6, 7, 18, 19]. The observed $M(H)$ dependence was reasonably well fitted to the Langevin function, therefore indicating the existence of weakly interacting Co particles. Above t^*_{Co}, the loops clearly exhibit a ferromagnetic contribution which saturates even at $H \sim 200$ Oe. We associate the occurrence of this ferromagnetic component with the formation of continuous Co layers. Therefore, t^*_{Co} signals the coalescence of the Co particles, and the anisotropic contribution is thus the magnetoresistive response of the continuous or quasicontinuous Co layers. This statement is firmly supported by the dependence of the RT thermal coefficient of resistivity (TCR), α, on t_{Co}. In figure 4, we have plotted α versus Co thickness (t_{Co}) at 290 K in multilayers of fixed Ag thickness, [Ag(20 Å)–Co(t_{Co})]$_{15}$. The TCR value α is defined by

$$\alpha = \frac{1}{\rho(290\ \mathrm{K})}\frac{\mathrm{d}\rho}{\mathrm{d}T}.$$

For a Co layer thickness of around 4.5 Å corresponding to the threshold value t^*_{Co}, this coefficient exhibits an abrupt increase. This observed discontinuity in α yields the thickness in which cobalt particles coalesce into continuous layers. The resistivity has a value of around 34 $\mu\Omega$ cm below t^*_{Co} and decreases up to in principle a value of around 28 $\mu\Omega$ cm for Co thickness of 10 Å. These values are in agreement with others previously reported for Co films [20]. The thermal dependence of the electrical resistivity for a series of [Ag(20 Å)–Co(t_{Co} Å)]$_{15}$ multilayers, $t_{\mathrm{Co}} = 2$, 4, 5, and 7, has been plotted in the inset of figure 4. A remarkable increase of the metallic regime is clearly displayed upon surpassing t^*_{Co} (plot c). This feature accords well with the above-mentioned cobalt coalescence, showing different behaviours below and above t^*_{Co}.

Measurements of MR loops (see figure 1(c)) at fields higher than 200 Oe show that the mechanisms responsible for the isotropic contribution for MR remain even for $t_{\mathrm{Co}} > t^*_{\mathrm{Co}}$. The isotropic magnetoresistance becomes progressively smaller as the rate of Co growth is increased in the multilayer. The isotropic component at higher fields is due to the spin-

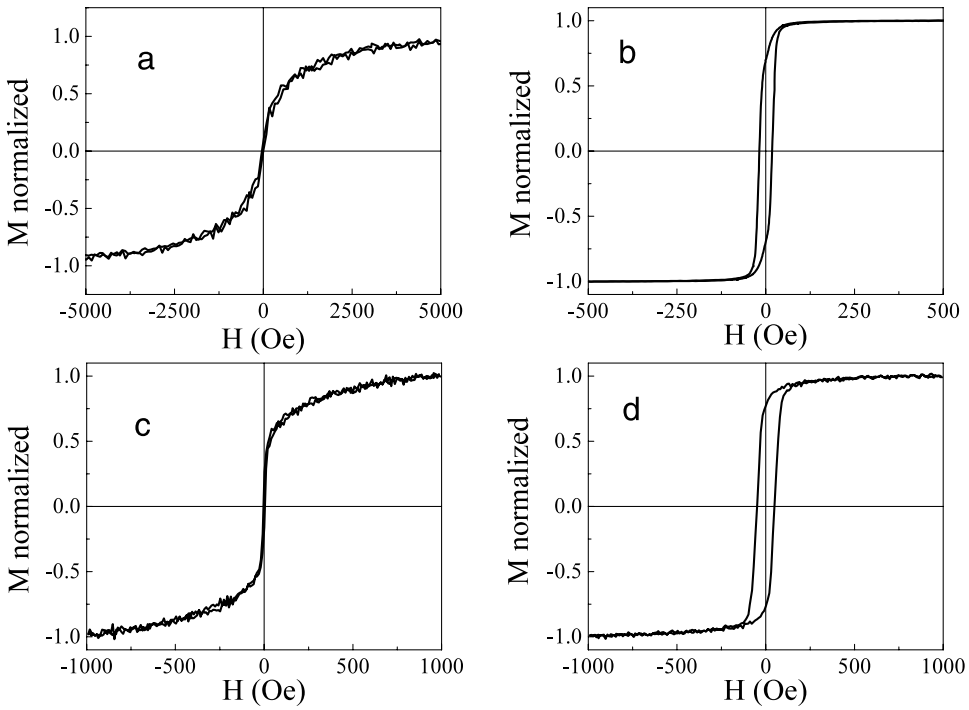

Figure 3. M–H hysteresis loops for series of [Ag(40 Å)–Co(t_{Co} Å)]$_{15}$ multilayers with (a) $t_{Co} = 5$ Å, (b) $t_{Co} = 60$ Å, and [Ag(10 Å)–Co(x Å)]$_{15}$ with (c) $t_{Co} = 4$ Å, and (d) $t_{Co} = 10$ Å.

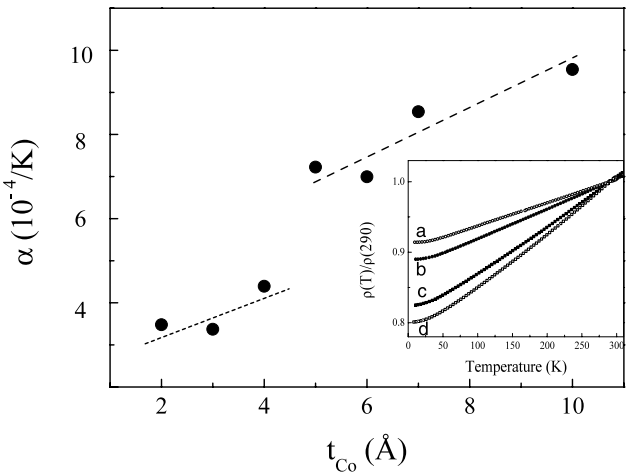

Figure 4. Temperature coefficient of resistivity, α, versus Co thickness in multilayers of fixed Ag thickness [Ag(20 Å)–Co(t_{Co} Å)]$_{15}$ at RT. Lines are drawn as a guide for the eyes. The inset shows the normalized resistance of four samples of the same series as a function of temperature. The cobalt layer thicknesses are (a) 2 Å, (b) 4 Å, (c) 5 Å, and (d) 7 Å.

dependent scattering at the Co cluster surfaces, whereas the anisotropic component is related to the spin-dependent scattering of conduction electrons within Co layers as well as at interfaces between Co and Ag layers [5, 9, 18].

For all the samples, the MR does not depend significantly on the relative orientation of the field and current at high fields. The field dependence of the MR measured up to 15 kOe has been found to be very sensitive to t_{Co}. The study of the MR dependence on t_{Co} has uncovered an important new physical aspect of the Ag/Co multilayers. Figure 5 shows the field dependence of the TMR for [Ag(20 Å)–Co(t_{Co})]$_{15}$ multilayers with constant $t_{Ag} = 20$ Å and variable Co thicknesses of 3, 4, 6, 7, and 20 Å. In this figure, the important influence of t_{Co} can be noted. Initially, TMR increases with Co thickness, reaching the maximum value of around 6% for 4 Å, and significantly decreasing for higher t_{Co}. To investigate whether t_{Ag} affects the evolution of the TMR as a function of t_{Co}, we have studied three series with constant Ag layer thickness and variable Co layer thickness. In figure 6, we plot the maximum value of the MR, defined as $MR_{max} = |MR\%(H = 1.5\ T)|$, versus t_{Co} for three different series with $t_{Ag} = 10$, 20, and 30 Å. MR_{max} represents the maximum increase of MR% obtained at a field of 1.5 T. These MR_{max} curves reach a clear maximum when varying t_{Co} in each of the series studied. This maximum is shifted to higher Co thickness as t_{Ag} is increased. In this plot, the largest values of MR_{max}, considered indeed as GMR, are found in the series with the smallest value of t_{Ag}. The MR loops, for low t_{Co}, displayed in figure 5, show a parabolic-like dependence on H without a noticeable hysteresis. That would suggest that multilayers with t_{Co} below the maximum value of MR_{max} have a superparamagnetic character due to Co particles or clusters. The initial positive slope of the MR_{max} curves is probably due to the increasing density of magnetic Co particles and their diminishing relative distances. This feature is similar to the observed behaviour in granular thin films [6, 18]. As t_{Co} approaches the maximum, the magnetic clusters begin to connect with adjacent ones to form cluster networks. For higher t_{Co}, the Co coalescence process occurs and multidomain Co layers are formed. As continuous Co layers are formed, the density of small isolated Co particles decreases, causing a decrease in MR_{max} because of the small contribution of ferromagnetic multidomains to the MR [9, 18]. This could explain the evolution of MR_{max} for $t_{Co} > t^*_{Co}$. Some authors have previously suggested, for different systems, that residual magnetic particles responsible for the magnetoresistance response are located near the interfaces [5, 21, 22]. Another possibility is that those particles could form within the volume of the Ag layers, similar to what occurs in the low-miscibility Ag–Co granular alloys. In order to investigate this explanation, MR loops have been measured in a series of Ag/Co multilayers with constant Co thickness ($t_{Co} > t^*_{Co}$) and varying t_{Ag} over a broad range. The results for MR_{max} versus t_{Ag} shown in figure 7 reveal a clearly linear increase up to roughly 40 Å, and a saturation regime being reached for higher t_{Ag}. This suggests that the rugosity of these interfaces increases linearly with t_{Ag} up to around 40 Å. To corroborate this statement we have performed low-angle x-ray reflectivity scans for samples with different values of t_{Ag}. The samples exhibit an interface roughness which increases with increasing t_{Ag}. This agrees with previous work concerning roughness in multilayers, where magnetoresistance was enhanced by the interface rugosity [23]. As an example, figure 8 shows LAXR results for a set of [Ag(t_{Ag} Å)–Co(10 Å)]$_{15}$ multilayers. The curves in those figures are the fits to the data which have been calculated with the REFS MERCURY refinement program. The evolution of the roughness of the silver layer versus t_{Ag} (the inset of figure 8) displays a behaviour similar to that of MR_{max} (figure 7). As we explained above, the isotropic magnetoresistance remains significant after the cobalt coalescence. This implies the presence of Co particles even after a continuous Co layer is formed. We suggest that residual Co particles must be located within the volume of the Ag layers. To explain the formation of Co particles inside the Ag layers, one must take into account the clustered growth of the Ag layer. Thus, grain boundaries in Ag could act as diffusion channels through which Co atoms penetrate well inside the spacers. Diffusion into the Ag grains is inhibited by the negligible Ag–Co solubility,

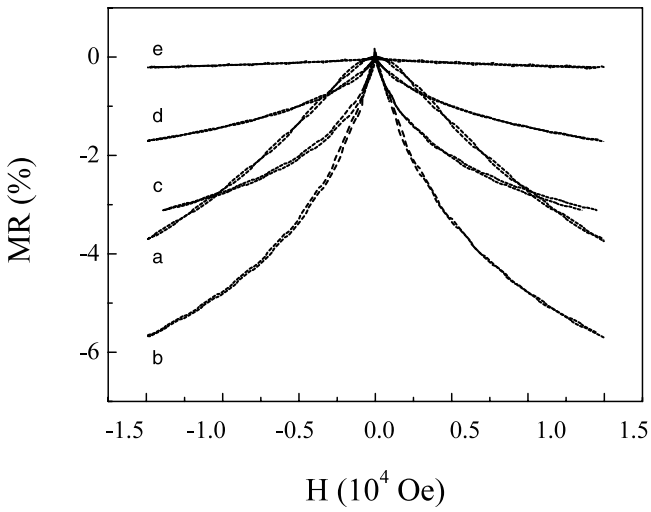

Figure 5. Magnetoresistance (MR) curves versus applied magnetic field for [Ag(20 Å)–Co(t_{Co} Å)]$_{15}$ multilayers: (a) $t_{Co} = 3$ Å, (b) $t_{Co} = 4$ Å, (c) $t_{Co} = 6$ Å, (d) $t_{Co} = 7$ Å, and (e) $t_{Co} = 20$ Å. The magnetic field is applied perpendicular to the electric current.

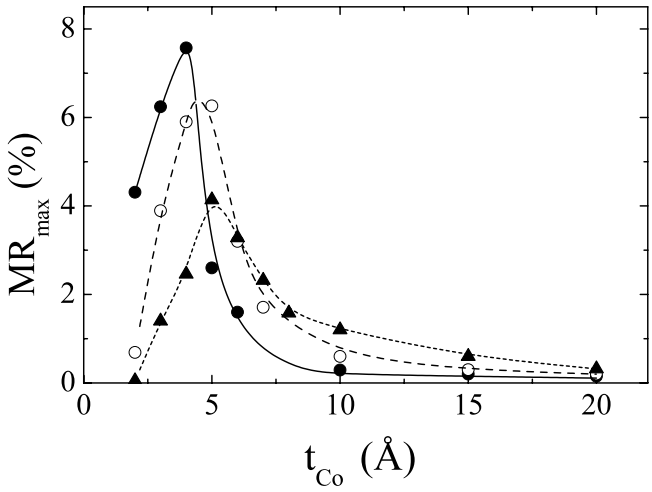

Figure 6. The dependence of the absolute value of MR% (1.5 T) on the cobalt thickness, t_{Co}, for three series of Ag/Co multilayers: $t_{Ag} = 10$ Å (solid circles), $t_{Ag} = 20$ Å (open circles), and $t_{Ag} = 30$ Å (triangles). Solids curves are drawn as a guide for the eyes.

and, therefore, the Co atoms would precipitate in favourable places among the Ag clusters or at grain boundaries.

4. Summary

We have found two different contributions in the magnetoresistance of the Ag/Co multilayers at low fields (<100 Oe): isotropic magnetoresistance for multilayers with Co thickness lower than around 5 Å, and anisotropic ones for multilayers with higher thicknesses. The transition

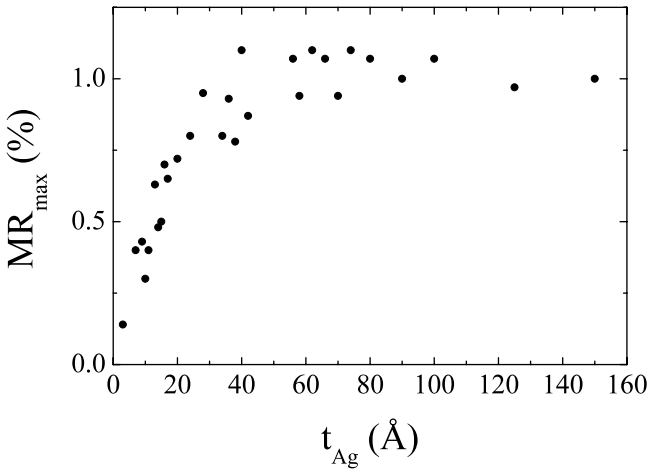

Figure 7. The dependence of the absolute value of MR% (1.5 T) on the silver layer thickness for a set of [Ag(t_{Ag} Å)–Co(10 Å)]$_{15}$ multilayers.

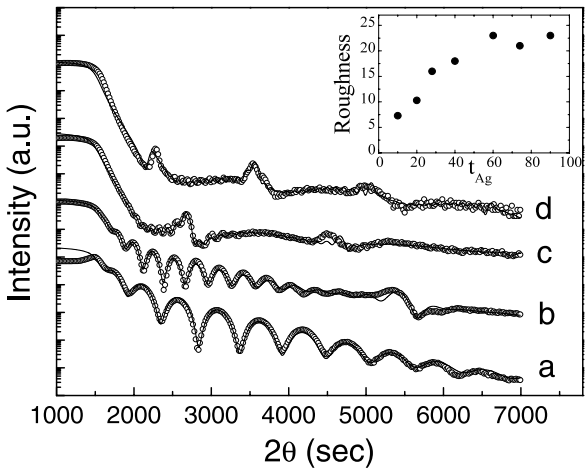

Figure 8. Low-angle x-ray reflectivity scans for a set of [Ag(t_{Ag} Å)–Co(10 Å)]$_{15}$ multilayers: (a) $t_{Ag} = 10$ Å, (b) $t_{Ag} = 20$ Å, (c) $t_{Ag} = 60$ Å, and (d) $t_{Ag} = 90$ Å. Curves are the fits obtained using the REFS MERCURY refinement program. The curves for different samples are displaced arbitrarily along the vertical axis for convenient graphical display. The inset shows the evolution of the roughness of silver versus t_{Ag} in the [Ag(t_{Ag} Å)–Co(10 Å)]$_{15}$ series.

from isotropic to anisotropic magnetoresistance, at low fields, takes place at a critical thickness t^*_{Co} which indicates the transition from a granular to a continuous magnetic layer. At t^*_{Co} the magnetization loops also change appreciably from isotropic granular superparamagnetic (below) to anisotropic layered ferromagnetic behaviour (above). The mechanisms responsible for the isotropic contribution persist even for $t_{Co} > t^*_{Co}$, although weakened, as the amount of Co in the multilayer increases, indicating that there are Co particles near the interfaces and within the volume of the Ag layers.

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