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Ferromagnetic resonance of ultrathin Co/Ag superlattices on Si(111)

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Ferromagnetic resonance (FMR) is used to probe the magnetic properties of Co/Ag superlattices (SLs) with ultrathin Co layers (2–6 Å). Different series of $5\times$[Ag/Co] multilayers have been grown by molecular beam epitaxy on Si(111) substrates, monitoring the growth by reflection high energy electron diffraction. Cross-section transmission electron microscopy confirms the growth of local areas with the designed SL periodicity, a sharp compositional modulation, well defined Ag–Co interfaces, and a perfect fcc (111) stacking. FMR spectra have been recorded at various polar angles in the 0°–90° range. A single and extremely broad resonance peak is observed in all cases. While SLs with Ag layers thinner than 10 Å exhibit similar values of the perpendicular anisotropy, a clear reduction is observed for samples with Ag layers about 14 Å thick. Possible causes for this change are discussed. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2839287]

I. INTRODUCTION

Layered films and superlattices (SLs) combining ferromagnetic (FM) and nonferromagnetic (NM) metals are challenging systems in order to provide materials with new specific properties. A recent example is the case of [Fe–Co/Pt] superlattices, proposed as a way of artificially manufacturing Fe–Co alloys with giant perpendicular magnetic anisotropy.[1] The importance of Si-based technologies makes attractive the growth of magnetic structures on single-crystal Si substrates. We analyze the microstructure and magnetic properties of [Co/Ag] SLs grown by molecular beam epitaxy (MBE) on Si(111) wafers. The Co/Ag system is rather singular and still controversial.[2–5] Achieving Co/Ag multilayers with flat, laterally continuous, and compositionally abrupt interfaces is particularly difficult.[2–5] Giant magnetoresistance (MR) values have been reported but oscillations of MR and magnetic coupling were only found for multilayers with Co layers <10 Å thick.[2,3] Perpendicular magnetic anisotropy has been reported for Co/Ag SLs, but again only for ultrathin Co layers.[4] Our Co/Ag SLs are analyzed by ferromagnetic resonance (FMR), a powerful tool to determine the contributions from different magnetic anisotropy fields. Recently, it has also been demonstrated its effectiveness in studying the exchange coupling in FM/NM/FM trilayers.[6]

II. SAMPLE GROWTH AND MICROSTRUCTURE

Samples are grown by MBE on nominally singular Si(111) substrates.[7] The multichamber MBE apparatus[8] is equipped with reflection high energy electron diffraction (RHEED) and Auger electron spectroscopy techniques. Clean 7×7-Si(111) surfaces are achieved by radiative heating (up to the temperature at which the 7×7 reconstruction appears $T_{R-7\times7}$). Film growth consists of successive Si(111) and Ag(111) buffer layers followed by five repetitions of the [Co/Ag] sequence (see Fig. 1). Further details can be found in Refs. 7 and 8. Samples are routinely characterized by x-ray diffraction (XRD),[8] and a few of them were also studied by transmission electron microscopy (TEM). Cross section samples for TEM were prepared by conventional methods and images were taken in a Philips CM200 with a field emission electron gun working at 200 kV.

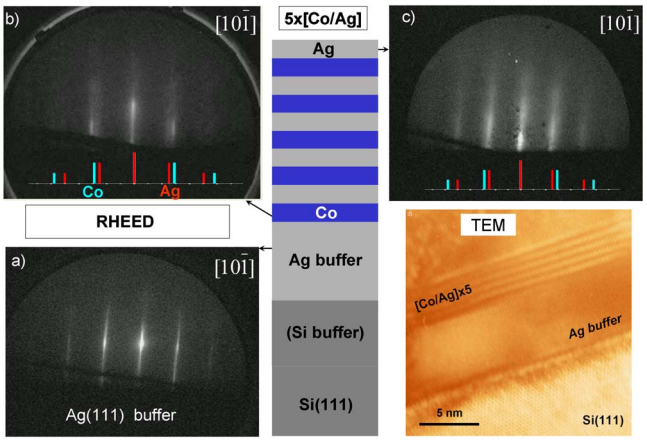

FIG. 1. (Color online) Sketch of layer growth sequence for the superlattice (SL) samples considered in this study (central part of the figure). RHEED patterns with the electron beam along the [1 0 −1] direction; they are representative of the stages indicated in the central sketch, corresponding to (a) completion of the Ag buffer layer, (b) growth of the first Co layer of the SL, and (c) growth of the last Ag layer of the SL. In the lower part of (b) and (c), we have indicated the diffraction rod positions for bulk Ag and Co under our experimental geometry. (d) Cross section TEM micrograph of a SL sample illustrating growth sequence; regions from the [Co/Ag] superlattice, Ag buffer layer, and Si buffer layer can be distinguished from top to bottom.

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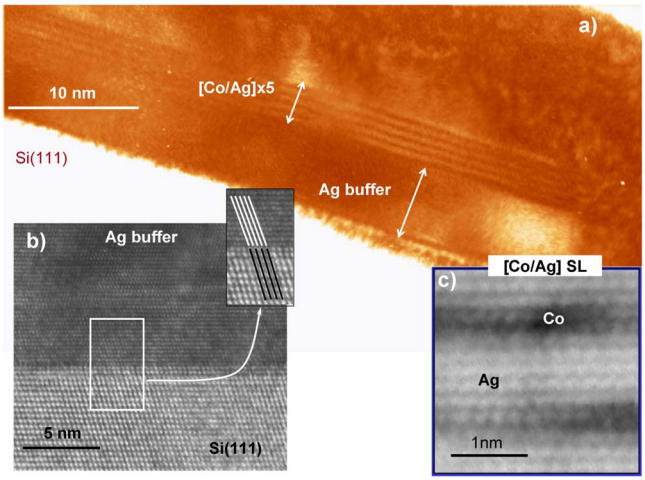

FIG. 2. (Color online) Cross section TEM images of two SL samples: picture (a) corresponds to a $5\times[\mathrm{Ag}(t_{\mathrm{Ag}})/\mathrm{Co}(t_{\mathrm{Co}})]$ SL with $t_{\mathrm{Ag}}=2$ ML and $t_{\mathrm{Co}}=1$ ML, while (b) and (c) display high resolution TEM micrographs of a SL with thicker Co layers ($t_{\mathrm{Co}}\approx 2-3$ ML). (b) Close-up view of the Ag buffer/Si buffer interface region; four Ag atomic rows match three atomic rows of Si (see inset), in accordance to the mismatch of 25% between bulk lattices. (c) Detailed view of two periods of the SL region, illustrating the fcc stacking of both Ag and Co layers.

The 2000 Å thick Si homoepitaxial layer is grown at a Si rate of 0.8 Å s^{-1} and substrate temperatures (T_{su}) close to $T_{R-7\times7}$. This procedure ensures a very clean and atomically flat Si(111)7×7 surface,[8] on which Ag buffer layers—with a thickness between 60 and 200 Å—are grown. Silver deposition is performed at T_{su}=RT using rates of either 0.5 or 0.05 Å s^{-1}. By inserting this Ag film we try to improve the crystallinity of the Co layers and to avoid the formation of Co silicides. The RHEED pattern of Fig. 1(a) illustrates the high crystalline quality of the Ag film, while the TEM images of Figs. 2(a) and 2(b) demonstrate the atomically flat and extremely abrupt Ag/Si interfaces achieved. Silver grows adopting its natural lattice constant from the very interface [Fig. 2(b)].

Different series of $5\times[\mathrm{Ag}(t_{\mathrm{Ag}})/\mathrm{Co}(t_{\mathrm{Co}})]$ multilayers have been fabricated at T_{su}=RT, alternating Co and Ag depositions. Cobalt layers are grown at 0.05 Å s^{-1} and Ag layers at the same rates as the buffer film. The thickness of Co layers (t_{Co}) is kept constant in each of the series, while that of Ag layers (t_{Ag}) is varied from about 3 to 15 Å. Only SLs with ultrathin Co layers (2–6 Å) are considered here. Growth is followed in real time by RHEED. The positions of the diffraction rods accurately correspond to Ag and Co bulk lattice parameters [Figs. 1(b) and 1(c)], indicating that Co and Ag layers grow keeping their own bulk lattice constants, in agreement with previous results.[4] The Co–Ag interfaces of the SL are thus incoherent, and each of the layers is presumably strain-free. Cross-section TEM micrographs confirm the presence of Co/Ag SL layers on various sample areas, and for different sets of t_{Ag} and t_{Co}; see Figs. 2(a) and 2(c) for $t_{\mathrm{Co}}=1$ atomic monolayer (ML) and $t_{\mathrm{Co}}=2-3$ ML, respectively. A sharp compositional modulation and well defined Co/Ag interfaces are observed in the SL regions. TEM images reveal a well ordered fcc atomic stacking of both Co and Ag SL layers [Fig. 2(c)], with ⟨111⟩ growth direction; fcc stacking was also found[4] for Co layers (few monolayer thick) in Co/Cu(111) and Co/Ag(111) SLs grown at $T_{\mathrm{su}}<323$ K. On the other hand, problems such as the lack of lateral continuity of the SL films are not solved yet, and we even detect areas with no trace of SL formation in one of the samples examined by TEM, Co hcp grains, and mixed Ag–Co regions appearing instead. Note, nevertheless, that the SL films shown in the TEM micrographs of Figs. 1 and 2 are among the best reported for Co/Ag multilayers and indicate a high degree of success in the growth of this difficult system.

III. MAGNETIC PROPERTIES

FMR measurements were done at RT and 9.85 GHz using an *X*-band electron paramagnetic resonance spectrometer Bruker ESP 300 E. The resonance field $H_r(\theta)$ was measured at different polar angles θ from $\theta=0°$ to $\theta=90°$. Data were processed using the Kittel equation with two fitting parameters: the *g*-factor (varying in the range of 2–2.25 for ferromagnets) and the effective anisotropy field H_{eff} that can be written as $H_{\mathrm{eff}}=4\pi M_s-H_{\perp}$, where M_s is the saturation magnetization and $H_{\perp}$ is a sum of all possible perpendicular anisotropy fields.[9] In the case of FM/NM multilayers, the most significant contributions are the surface (H_s), magnetocrystalline (H_k), and magnetoelastic (H_σ) anisotropies, and that caused by the indirect exchange between neighbor layers (H_{ex}). We measured first a 300 Å thick Co film grown on a 200 Å thick Ag buffer. The best fit of its angular dependence $H_r(\theta)$ was obtained for an effective anisotropy field $H_{\mathrm{eff}}=17.3$ kOe and a *g*-factor=2.1, which are typical values of polycrystalline Co.

Three [Co/Ag] SLs are selected to discuss the FMR results: they have a fixed Co thickness ($t_{\mathrm{Co}}\approx 4$ Å) and different Ag thicknesses ($t_{\mathrm{Ag}}\approx 4.5$, 9, and 13.5 Å, respectively). Only one and very broad resonance peak (linewidth about 2 kOe) is observed in the spectra of the three samples (see Figs. 3 and 4). For the SLs with $t_{\mathrm{Ag}}\approx 4.5$ and 9 Å, we find almost identical values of the perpendicular anisotropy $H_{\perp}$ (9.1 and 9.2 kOe, respectively), with a similar *g*-factor=2.07; results displayed in Fig. 3 correspond to the SL with $t_{\mathrm{Ag}}\approx 4.5$ Å. In contrast, for the SL sample with $t_{\mathrm{Ag}}\approx 13.5$ Å (Fig. 4), a much lower value ($H_{\perp}\approx 4$ kOe) is found. The FMR measurements thus indicate a significant reduction (50%) of $H_{\perp}$ for the SLs with $t_{\mathrm{Ag}}\approx 13.5$ Å, compared to those SLs with $t_{\mathrm{Ag}}<10$ Å. Bearing in mind the results on sample microstructure, it seems reasonable to assume that the H_s, H_k, and H_σ contributions would be rather similar in the three SLs grown upon similar conditions and with the same t_{Co}. Hence, the most probable candidate to explain the observed differences in $H_{\perp}$ seems to be H_{ex}. To clarify the nature of interlayer interactions, magneto-optic Kerr effect (MOKE) and vibrating sample magnetometry (VSM) measurements have been additionally performed on the SL samples of Figs. 3 and 4. The $M(H)$ curves recorded at RT by VSM and MOKE techniques indicate a typical ferromagnetic behavior and a rather high coercive field H_c for both samples, ranging from $H_c=400$ Oe ($t_{\mathrm{Ag}}=4.5$ Å) to $H_c=415$ Oe ($t_{\mathrm{Ag}}=13.5$ Å) (see insets of Figs. 3 and 4). In case of ferromagnetic coupling, an increase of the interaction strength should shift the position

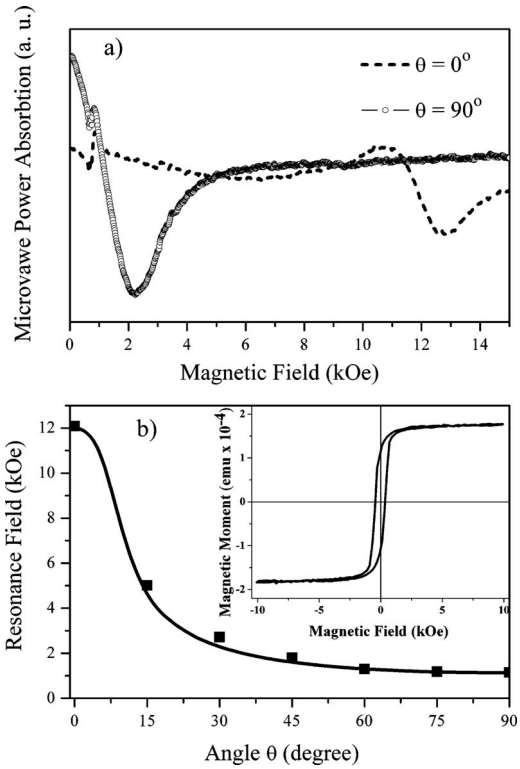

FIG. 3. Results of FMR and VSM measurements at RT for a $5\times$[Co(4 Å)/Ag(4.5 Å)] SL sample. (a) Ferromagnetic resonance signals at two polar angles: $\theta=0°$ ($\boldsymbol{H}\|\mathbf{n}$) and $\theta=90°$ ($\boldsymbol{H}\perp\mathbf{n}$), where $\boldsymbol{H}$ is the external applied field and $\mathbf{n}$ the normal vector to the film plane. (b) Out-of-plane angular dependence $H_r(\theta)$. Dots are experimental data, while solid lines represent the theoretical fit. Fitting parameters are $H_{\text{eff}}=7.83$ kOe and $g=2.07$. The in-plane magnetization loop measured by VSM is shown in the inset.

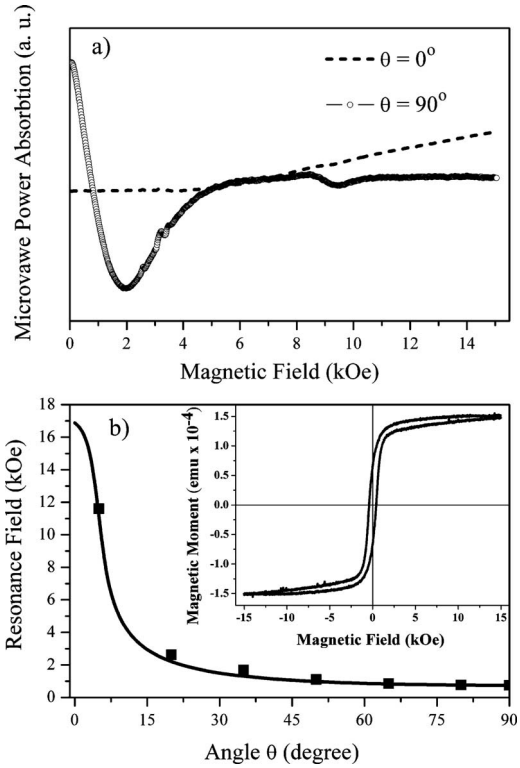

FIG. 4. Same as Fig. 3, for the case of a $5\times$[Co(4 Å)/Ag(13.5 Å)] superlattice. (a) Note that in perpendicular geometry, $\theta=0°$ ($\boldsymbol{H}\|\mathbf{n}$), the resonance field is higher than the maximum spectrometer field. (b) Fitting parameters are $H_{\text{eff}}=13.6$ kOe and $g=2.1$.

of the in-plane resonance peak toward lower fields,[6] situation actually found in Fig. 4. Thus, the 50% reduction of $H_\perp$ observed for the SL with $t_{\text{Ag}}\approx 13.5$ Å, in comparison to SLs with $t_{\text{Ag}}<10$ Å, seems to reflect an increase of the interlayer coupling. Among the rest of contributions to perpendicular anisotropy, H_s is expected to be the most significant one. Indeed, H_s was reported[4] to be large enough to change the sign of H_{eff} in Co/Ag multilayers when $t_{\text{Co}}\leqslant 3$ Å. However, here we should not expect large variations in its contribution to H_{eff} in view of the similar high quality of the Co/Ag interfaces revealed by RHEED and TEM. We would not either expect a strong influence of H_k on the reduction of $H_\perp$ for $t_{\text{Ag}}\approx 13.5$ Å since the Co layers of our SLs exhibit similar fcc (111) crystal structures. No trace of in-plane anisotropy was found in MOKE measurements when samples were rotated in external field, which also suggests a small H_k contribution. Finally, the magnetoelastic anisotropy (H_σ) should be negligible since Co layers in the SLs are almost strain-free, as shown by RHEED.

In conclusion, Co/Ag superlattices with individual Co layer thickness in the range from 1 to 3 ML have been fabricated by molecular beam epitaxy on Si(111) substrates. For SLs with 2 ML thick Co layers and Ag layer thickness between 4 and 14 Å, FMR reveals the existence of significant perpendicular magnetic anisotropy at RT. A reduction of nearly 50% in the perpendicular anisotropy is observed for SLs with Ag layers about 14 Å thick, as compared to samples with thinner Ag layers.

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