Magnetic properties of amorphous $Co_{0.74}Si_{0.26}$/Si multilayers with different numbers of periods

G. N. Kakazei[a)]

IFIMUP and IN-Institute of Nanoscience and Nanotechnology, Departamento de Física da Faculdade de Ciências, Universidade do Porto, Porto 4169-007, Portugal, Departamento de Física, Universidad de Oviedo, Oviedo 33007, Spain, and Institute of Magnetism of the National Academy of Sciences of Ukraine, 36b Vernadskogo Blvd., Kiev 03142, Ukraine

N. M. Santos

I3N and Departamento de Física, Universidade de Aveiro, Aveiro 3810-193, Portugal

C. Quiros, M. Velez, J. I. Martin, and J. M. Alameda

Departamento de Física, Universidad de Oviedo, Oviedo 33007, Spain

V. O. Golub and O. Y. Saliuk

Institute of Magnetism of the National Academy of Sciences of Ukraine, 36b Vernadskogo Blvd., Kiev 03142, Ukraine

Yu. G. Pogorelov and J. B. Sousa

IFIMUP and IN-Institute of Nanoscience and Nanotechnology, Departamento de Física da Faculdade de Ciências, Universidade do Porto, Porto 4169-007, Portugal

M. C. Carmo and N. A. Sobolev

I3N and Departamento de Física, Universidade de Aveiro, Aveiro 3810-193, Portugal

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Two sets of $[Co_{0.74}Si_{0.26}(5\ nm)/Si(s)]_n$ amorphous films were prepared by magnetron sputtering: one in the form of multilayers with the Si spacer thickness s fixed at 3 nm, and the number n of periods varying from 1 to 10 and the other with only two periods and s varying from 3 to 24 nm (trilayers). In both sets, the $Co_{0.74}Si_{0.26}$ layer thickness t was fixed at 5 nm. All the samples except the one with $s=24$ nm manifest antiferromagnetic coupling. Their magnetic properties at room temperature were probed using the magnetooptical transverse Kerr effect (MOTKE) and ferromagnetic resonance (FMR). The relative increase in the saturation magnetization M_s (for trilayers, relative to a structure with $s=24$ nm; for multilayers, relative to the single-layer structure) determined from the FMR measurements was compared with the exchange coupling strength H_J^{AF} obtained from the MOTKE studies. The dependences of H_J^{AF} and M_s on n and s were found to be very similar to each other. Possible mechanisms of this similarity are discussed. © *2010 American Institute of Physics*. [doi:10.1063/1.3499251]

I. INTRODUCTION

Interlayer exchange coupling (IEC) through a silicon spacer was discovered in Fe/Si multilayers in 1992.[1] Since that time different ferromagnetic metal (FM)/Si multilayers have been studied experimentally and theoretically. Many of these studies have dealt with Fe/Si multilayers, where the coupling shows both oscillatory[1] and nonoscillatory[2] behavior and is strongly influenced by the interface structure (namely the formation of iron silicide owing to diffusion).[3,4] Even more contradictory results were obtained for Co/Si structures, where ferromagnetic, superparamagnetic, or oscillatory ferromagnetic-antiferromagnetic behavior of the coupling has been observed.[5–8] Although several mechanisms based on tunneling, Ruderman–Kittel–Kasuya–Yosida (RKKY)-like exchange, interface bands, and spin fluctuations have been suggested as the cause of the coupling, there is still little theoretical understanding of these systems.

Recently antiferromagnetic (AF) coupling has been discovered in multilayers based on amorphous Co_xSi_{1-x} alloys.[9–11] One characteristic of these magnetic amorphous compounds, besides their well known soft magnetic behavior, is that several properties, such as the saturation magnetization and uniaxial anisotropy, can be tuned by fine adjustment of the alloy composition. In the case of Co_xSi_{1-x}/Si multilayers, the saturation magnetization and the Curie temperature are reduced when the Si content is increased,[10] so that the strength of the usual magnetostatic coupling contributions present in any magnetic multilayer system can be tailored. Moreover, the soft magnetic behavior of these alloys allows the detection of very weak AF coupling which could not be observed in samples based on pure Co magnetic layers. It is known from previous studies that Co_xSi_{1-x} films are amorphous for Co concentrations smaller than $x=0.76$ and have low coercive fields (below 1 Oe for 5 nm thick films).[12] Thus, $Co_{0.74}Si_{0.26}$/Si multilayers appear to be a good choice of a system for studying weak AF coupling, since they have relatively soft magnetic behavior which enables the detection of a plateau on the $M(H)$ hysteresis loop around $H=0$, a clear footprint of AF coupling. It is also important that the AF coupling strength can be easily derived from the $M(H)$ loop.

Ferromagnetic resonance (FMR) has an established reputation as a powerful tool for investigating the magnetic parameters of thin films, multilayers, and patterned structures; it is particularly useful for determining the contributions from different magnetic anisotropy fields. Its effectiveness has also been demonstrated in studies of exchange coupling in ferromagnetic metal (FM)/nonferromagnetic metal (NM)/FM trilayers.[13] Here the evolution of the FMR resonance field in $\mathrm{Co_{0.74}Si_{0.26}/Si}$ multilayers has been studied as a function of AF interlayer exchange strength, which is controlled either by varying the Si layer thickness or the number of periods.

II. EXPERIMENTAL DETAILS

$\mathrm{Co_{0.74}Si_{0.26}/Si}$ multilayers were grown on Si substrates by dc magnetron sputtering from independent high purity Co and Si targets. The sputtering pressure was $1.0\cdot 10^{-3}$ mbar (with a base pressure of $\sim 10^{-9}$ mbar). The Co target was placed at normal incidence with respect to the substrate, while the Si atoms arrive at oblique incidence ($\sim 30°$ with respect to the substrate normal). Two series of $[\mathrm{Co_{0.74}Si_{0.26}}(t)/\mathrm{Si}(s)]_n$ multilayers were prepared: one with the Si spacer thickness s fixed at 3 nm, and the number of periods n varying from 1 to 10, and another with only two periods and s varying from 3 to 24 nm. In both series the $\mathrm{Co_{0.74}Si_{0.26}}$ layer thickness t was fixed at 5 nm. In all cases a 3 nm thick Si buffer layer was grown on top of the native oxide of the substrate before growing the corresponding multilayer. A protective Si capping layer of the same thickness was always deposited on top of the samples in order to prevent oxidation.

A magnetooptical transverse Kerr effect (MOTKE) system was used to study the hysteresis loops of the samples at room temperature. The MOTKE signal, δK, is defined as $\delta K=(R^{+}-R^{-})/R$, where R^{+} is the reflectivity for a positive applied magnetic field, R^{-} is that for a negative field, and R is the value for an idealized nonmagnetized sample, taken, in practice, to be the average of R^{+} and R^{-}. For a thin film, δK is proportional to the magnetooptic constant Q which, in a first approximation, is linear in the saturation magnetization M_S.

The $\mathrm{Co_{0.74}Si_{0.26}}(t)/\mathrm{Si}$ multilayers were probed by continuous wave ferromagnetic resonance. The FMR field at room temperature was measured using a standard Bruker ESP 300 (~ 9.8 GHz) X-band electron paramagnetic resonance spectrometer for the full range of angles θ between the external field direction and the normal to the film plane (0°–90°). The in-plane angular dependences (azimuthal angle φ ranging from 0° to 360° for $\theta=90°$) of the resonance field H_r were also measured to determine the in-plane uniaxial anisotropy, which is to be expected in this system according to previous studies.[9,10]

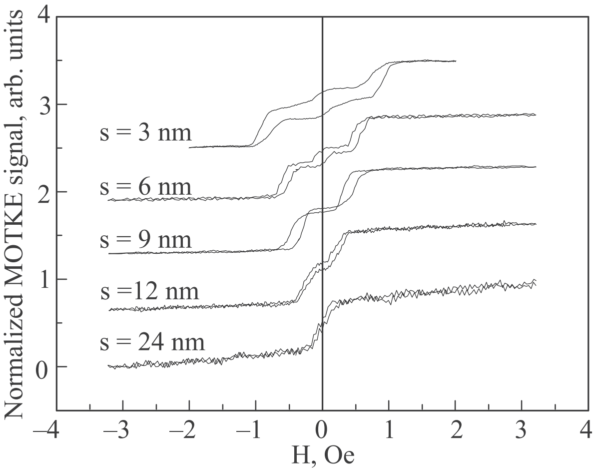

FIG. 1. Normalized MOTKE hysteresis loops for $[\mathrm{Co_{0.74}Si_{0.26}}(5\ \mathrm{nm})/\mathrm{Si}(s)]_2$ with different thicknesses of the Si layer s. The curves have been shifted vertically for clarity.

III. RESULTS AND DISCUSSION

The magnetooptical studies revealed that a single layer of $\mathrm{Co_{0.74}Si_{0.26}/Si}$ has a coercive field of 0.5 Oe, and has, as do all the other multilayered samples involved in the experiment, a well defined uniaxial anisotropy with an anisotropy field of approximately 20 Oe. The magnetic behavior of the series of $\mathrm{Co_{0.74}Si_{0.26}}$ (5 nm)/Si(s nm)/$\mathrm{Co_{0.74}Si_{0.26}}$ (5 nm) trilayers is illustrated in Fig. 1 which reveals the clear presence of AF coupling for Si spacer thicknesses up to 12 nm. All the samples except the one with $s=24$ nm have an almost zero remanence, yielding a clear plateau around $H=0$. A step in the loop appears at magnetization values close to zero; this is related to the AF state of the trilayer. It can easily be seen that the coupling between the layers falls off monotonically with increasing s and completely disappears above 20 nm. The coupling field strength H_J^{AF} for the trilayer samples was determined using the simple formula (proposed in Ref. 10)

$$H_J^{AF}=\frac{H_3-H_1}{2}, \tag{1}$$

where H_1 is the field at which the reversal process initiates formation of an AF coupled state as the external field decreases continuously from its maximum positive value, and H_3 is the field where the AF state suddenly breaks up, leading to a rapid shift in the magnetization to values close to negative saturation.

The magnetic evolution of the MOTKE hysteresis loop as the number of periods increases in $[\mathrm{Co_{0.74}Si_{0.26}}(5\ \mathrm{nm})/\mathrm{Si}(3\ \mathrm{nm})]_n$ (in this case the AF coupling ($s=3$ nm) is strongest) is illustrated in Fig. 2. As the number of periods is raised, other steps appear, although their width with respect to the field decreases continuously, and the entire process of magnetization reversal between the two saturated states becomes smoother, as shown in Fig. 2 for the case of $n=6$ (note that the saturation field is much smaller than the anisotropy field, 20 Oe, obtained from the completely closed hard-axis hysteresis loops). None of the samples have zero MOTKE signal at zero field, but most have values very close to zero. In our case of only even numbers of periods, the signal does not go to zero, even for

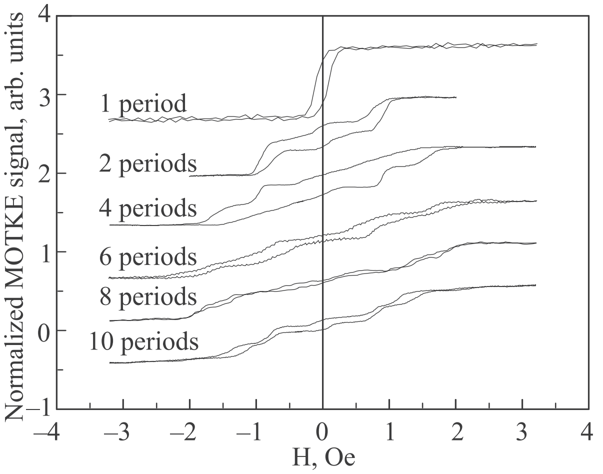

FIG. 2. Normalized MOTKE hysteresis loops for $[\mathrm{Co}_{0.74}\mathrm{Si}_{0.26}(5\ \mathrm{nm})/\mathrm{Si}(3\ \mathrm{nm})]_n$ with different numbers of periods. The curves have been shifted vertically for clarity.

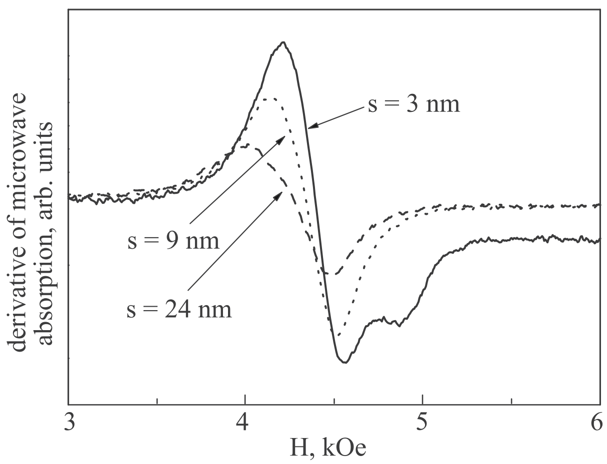

FIG. 3. Ferromagnetic resonance signals (first derivative of the microwave absorption) at $\theta=0°$ for $[\mathrm{Co}_{0.74}\mathrm{Si}_{0.26}(5\ \mathrm{nm})/\mathrm{Si}(s)]_2$ with different thicknesses s of the Si layer.

zero net magnetization, mainly because of interference effects. One can see that, as the number of periods increases, the strength of the interlayer interactions increases, saturating at $n=6-8$. For the multilayered samples the coupling field strength H_J^{AF} was derived from a formula similar to Eq. (1):

$$H_J^{AF}=\frac{H_4-H_1}{2}, \tag{2}$$

where H_4 is the field at which the magnetization reaches negative saturation. H_3 and H_4 are almost identical for the trilayers and it was easy to determine H_3 for them; however, only H_4 could be determined clearly for the multilayers.

The FMR data were fitted using the well-known Kittel equation with two parameters, the g-factor and the effective anisotropy field, $H_{\mathrm{eff}}=4\pi M_s-H_{\perp}$, where M_s is the saturation magnetization and $H_{\perp}$ is the sum of all possible perpendicular anisotropies (see Ref. 14 for more details). In the case of ferromagnetic/nonmagnetic multilayers, the expected contributions to H are as follows: surface related (H_s), magnetocrystalline (H_k) and magnetoelastic H_σ terms, plus one owing to indirect exchange between neighboring layers (H_{ex}).

In-plane FMR angular dependence measurements confirmed the presence of a weak in-plane uniaxial anisotropy in all the samples (also detected by MOTKE), with approximately the same anisotropy field value $H_{\text{in-plane}}\approx 20$ Oe. We note that, except in very special cases of strong in-plane uniaxial anisotropy (i.e., when $H_{\text{in-plane}}$ is comparable with resonance field; see, for example, Ref. 15), $H_{\text{in-plane}}$ can be obtained easily using Eq. (6) of Ref. 14. For the single period $\mathrm{Co}_{0.74}\mathrm{Si}_{0.26}$ film, the best fit to the out-of-plane $H_r(\theta)$ dependence was obtained with an effective anisotropy field $H_{\mathrm{eff}}\approx 0.9$ kOe corresponding to $M_s\simeq H_{\mathrm{eff}}/4\pi=73$ G, in good agreement with previous studies.[9,10] On going from a single layer film to a trilayer film ($n=2$) with a Si interlayer thickness $s=3$ nm, the effective anisotropy field increases by 100 Oe. With increasing Si interlayer thickness at $n=2$, the effective anisotropy decreases to the value for a single layer film $H_{\mathrm{eff}}\approx 0.9$ kOe at $s=24$ nm (see Fig. 3). For the second series (s fixed at 3 nm, n varying from 2 to 10), the effective anisotropy gradually increases up to 1050 Oe and saturates at $n=8$ (see Fig. 4).

The observed relative changes in the effective anisotropy (for trilayers relative to the structure with $s=24$ nm; for multilayers relative to the single layer structure) were compared with the exchange coupling strength H_J^{AF} (see Figs. 5 and 6, respectively). It can be seen that the variations in H_J^{AF} and H_{eff} are very similar to one another in each case. This suggests that, although it is very weak, exchange coupling has a significant influence on the effective anisotropy of the system: for the $[\mathrm{Co}_{0.74}\mathrm{Si}_{0.26}(5\ \mathrm{nm})/\mathrm{Si}(3\ \mathrm{nm})]_{10}$ multilayer structure the effective anisotropy is $\sim 17\%$ greater than for the single-layer film. There are several possible mechanisms that could change the effective anisotropy of the system. As mentioned above, the effective anisotropy includes shape anisotropy (proportional to the saturation magnetization), and surface-related (H_s), magnetocrystalline (H_k) and magnetoelastic (H_σ) contributions, as well as a contribution owing to indirect exchange between neighboring layers (H_{ex}). In the case of amorphous magnetic layers, H_k and H_σ should be negligibly small compared to $4\pi M_s$, while H_s for the given ferromagnetic layer thickness $t=5$ nm is also very small and

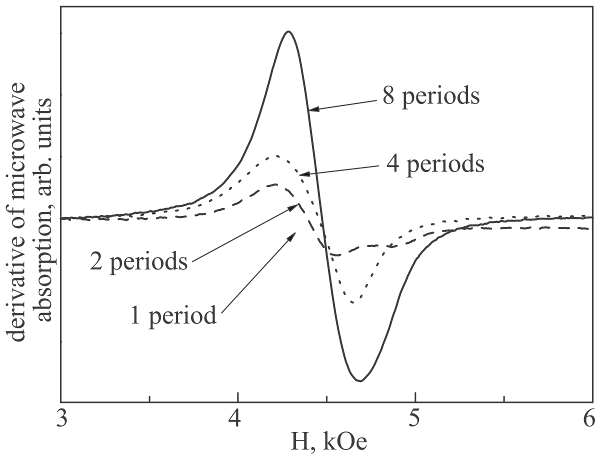

FIG. 4. Ferromagnetic resonance signals (first derivative of the microwave absorption) at $\theta=0°$ for $[\mathrm{Co}_{0.74}\mathrm{Si}_{0.26}(5\ \mathrm{nm})/\mathrm{Si}(3\ \mathrm{nm})]_{\mathrm{n}}$ with different numbers of periods.

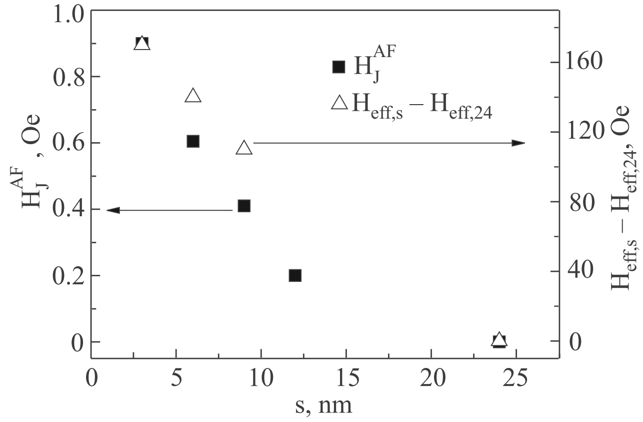

FIG. 5. Comparison of the exchange field strength (obtained from MOTKE measurements) and the change in the effective anisotropy field relative to the sample with $s=24$ nm (obtained from FMR measurements) for trilayers with different Si layer thicknesses.

independent of n, so that all these contributions can be ignored in the following analysis. Exchange coupling H_{ex} could also modify H_{eff}; however, direct calculations of the contribution from H_{ex} using the equations from Ref. 13 show that, even for the strongest coupling (i.e., the $\mathrm{Co}_{0.74}\mathrm{Si}_{0.26}(5\ \mathrm{nm})/\mathrm{Si}(3\ \mathrm{nm})]_{10}$ sample) the additional contribution to H_{eff} will be ~ 10 Oe, i.e., a factor of 17 below the value obtained from the FMR experiments. Furthermore, in case of AF exchange coupling strong enough to noticeably change the FMR resonance fields, the $H_r(\theta)$ angular dependence will change in a way different from that observed here: for both in-plane and perpendicular-to-plane configurations H_r will shift to higher fields than for a noninteracting sample. As a result, a formal analysis using the Kittel formula will yield a change in the g factor rather than in H_{eff}. Actually, the g factor was almost the same (≈ 2.15) for all the samples included in this study. Thus, the influence of H_{ex} on the effective anisotropy appears to be negligible.

Therefore, after all the other terms have been excluded, the only possible explanation left for the observed enhancement in H_{eff} is the variation in the saturation magnetization as a function of either the Si spacer thickness for the trilayers or of the number of periods for the multilayers. This change in M_s is proportional to the exchange coupling strength in the sample. Two possible factors could modify the room temperature M_s in these multilayers: First, varying the Si spacer thickness of the trilayer may change the Si diffusion rate from the spacer to the ferromagnetic layers, so that the Co content and magnetization of trilayers with different values of s will change gradually. However, this scheme cannot explain the gradual increase in the magnetization in the multilayered samples as the number of periods is increased. Also, in the case of interdiffusion, the Co concentration will gradually change from the edge to the center of a layer. This should increase the FMR linewidth of the samples with lower H_{eff} (i.e., saturation magnetization). However, no significant and systematic change of the FMR linewidth was found in the samples studied here. Second, it is known that the Curie point for $\mathrm{Co}_{0.74}\mathrm{Si}_{0.26}$ is close to room temperature. Hence, if exchange coupling can change T_C even slightly, it could cause a noticeable change in the room temperature magnetization which might explain all of the observed results for both series of samples.

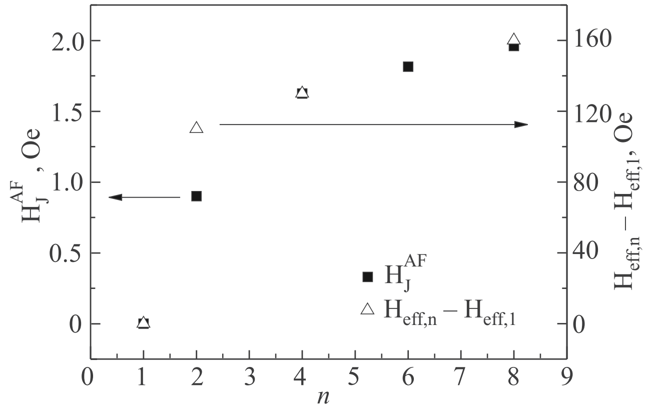

FIG. 6. Comparison of the exchange field strength (obtained from MOTKE measurements) and the change in the effective anisotropy field relative to that of a single-layer sample (obtained from FMR measurements) for multilayers with different numbers of periods.

CONCLUSIONS

The exchange coupling strength H_J^{AF} in $[\mathrm{Co}_{0.74}\mathrm{Si}_{0.26}(5\ \mathrm{nm})/\mathrm{Si}(s)]_n$ was tuned either by varying the Si spacer thickness s for the $[\mathrm{Co}_{0.74}\mathrm{Si}_{0.26}(5\ \mathrm{nm})/\mathrm{Si}(s)]_2$ series or by changing the number of periods n for the $[\mathrm{Co}_{0.74}\mathrm{Si}_{0.26}(5\ \mathrm{nm})/\mathrm{Si}(3\ \mathrm{nm})]_n$ series. The relative increase in the effective anisotropy field and in the corresponding saturation magnetization (for the trilayers relative to the structure with $s=24$ nm; for the multilayers relative to the single layer structure) obtained from the FMR measurements was found to be proportional to H_J^{AF}.

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[a] Email: gleb.kakazei@fc.up.pt

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