Frustration-driven micromagnetic structure in Fe/CoO/Fe thin film layered systems

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We have investigated the micromagnetic structure of magnetic domains in Fe/CoO/Fe trilayer systems and the magnetization coupling between the iron layers. We observe very small magnetic domains separated by nanometer-sized domain walls in the top Fe layer for a narrow CoO thickness range. Such domains have lateral dimensions as low as 30 nm and present topologies which are very similar to those observed in the top layer of Fe/NiO/Fe trilayers. Both magnetic domain structure and Fe interlayer coupling dramatically change with the CoO thickness. The role of magnetocrystalline anisotropy and magnetic frustrations on the observed phenomenology is discussed.

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The interest toward magnetic thin films multilayers has been quite lively so far, driven by the discovery of giant magnetoresistance and of its important applications in magnetoelectronic devices.[1] The focus of a significant part of the research in this field has been directed to unveil the mechanisms of interlayer exchange coupling (IEC) between ferromagnetic (FM) layers separated by a non-FM layer.[2,3] A particular case of non-FM spacer is that of an insulating and antiferromagnetic (AFM) thin film. This special case is characterized by the absence of Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling while exchange coupling between FM and AFM spins at the interfaces strongly influences the interactions between the FM layers.[2]

The behavior of FM/AFM multilayers depends on a large number of parameters, such as interface roughness, magnetic defects, and strain, which are known to induce frustrations in the magnetic couplings.[4,5] A significant indirect observation of the effects of frustrations at the FM/AFM interfaces is the occurrence of a biquadratic term in the IEC. Such an occurrence has been predicted to possibly lead a 90° in-plane coupling between the magnetizations of the FM layers.[2,3] This has eventually been observed in several cases.[4,6–8] Another recent prediction concerning the effects of frustrations suggests the presence of very small magnetic domains in FM films characterized in particular by very sharp domain walls (DW).[9] According to conventional theories applied to bulk materials, the magnetic domain structure results from the balance between magnetocrystalline anisotropy (MCA) and exchange forces. For thin layered systems, such as FM/AFM/FM multilayers, the model of Ref. 9 has underlined the role of magnetic frustrations as competitors in determining both domain structures and magnetic couplings.

Among insulating AFM materials, transition metal oxides such as CoO and NiO have been extensively employed in FM/AFM systems[10,11] and their structural and electronic properties are well known. Recently, we have observed very small magnetic domains in the top layer of Fe/NiO/Fe(001) systems.[12,13] Such domains have in-plane magnetization and lateral dimensions of the order of few tens of nanometers. The corresponding DW are extremely sharp, with thickness below 40 nm.

In this work, we investigate the magnetic properties of Fe/CoO/Fe(001) trilayer systems, focusing on the characteristics of the magnetic domains and DW in the top Fe layer and on their evolution as a function of the CoO thickness. By comparing systems characterized by similar interfaces but exhibiting large differences in the MCA values, it can be possible to shed light on the problem of disentangling among bulk (related to MCA) versus surface or interface (related to frustrations) effects in determining the magnetic properties of layered structures. In consideration of the very different values of the MCA constants in CoO, where $K_2=2\times10^5$ erg/cm^3, with respect to NiO, where $K_2=3.3\times10^2$ erg/cm^3,[14–16] the study of the magnetic domain structure of the top layer of Fe/CoO/Fe trilayers is extremely interesting. In the following we show, by means of x-ray photoelectron emission microscopy (X-PEEM), that nanometer-sized magnetic domains develop on the top Fe layer of our Fe/CoO/Fe trilayers, similarly as for the Fe/NiO/Fe case.

Fe/CoO/Fe samples have been prepared in ultrahigh vacuum as described in previous publications.[11,13,17] In particular, the CoO spacers have been grown onto the surface of a 100-nm-thick Fe(001) film through deposition of the metallic component in a controlled oxygen atmosphere and have thicknesses from 1 nm up to about 5 nm. The CoO thickness (t_{CoO}) has been varied continuously in a wedged fashion except for some plateaux of constant thickness. The latter have been subsequently used as references for the estimation of the thickness in the wedged regions. The upper Fe layer has a constant thickness of 3 nm. The samples have been exposed before the measurements to a magnetic field strong enough to induce saturation magnetization of the Fe substrate (see below). The experiments have been performed on the SOLEIL's X-PEEM microscope installed at the Nanospectroscopy beamline of the synchrotron ELETTRA, which features a 50 nm lateral resolution in the magnetic contrast mode.[18]

Imaging of the magnetic domains in Fe/CoO/Fe is obtained by collecting x-ray magnetic circular dichroism

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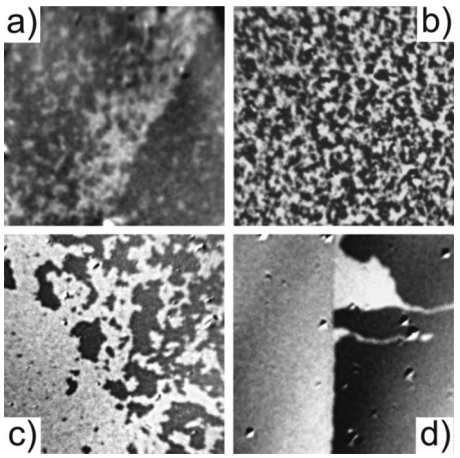

FIG. 1. Room-temperature XMCD asymmetry images from the top Fe layer in the Fe/CoO/Fe system. The CoO thickness is from panels (a) to (d): 1 nm, 2 nm, 4 nm, and 5 nm, respectively. The field of view is 10 μm $\times$ 10 μm for each image.

(XMCD) asymmetry images at the L_3 absorption edge of Fe, where the dichroic signal is maximum.[19,20] Figure 1 reports on the room-temperature evolution of the domains morphology in the top Fe layer with increasing t_{CoO}. The gray level in the images depends on the alignment between the propagation vector of the incident (circularly polarized) x rays and the sample magnetization vector. In the brighter domains, the two vectors are parallel to each other, while in the darker they are antiparallel. In the gray domains, such an alignment is between 0° and 180°. Figure 1(d) shows for instance three different gray levels for domains characterized by an in-plane magnetization vector either parallel, antiparallel, or perpendicular with respect to the light propagation vector.[21] The t_{CoO} value has been estimated by averaging the absorption signal at the Co $L_{2,3}$ edges over each image area and comparing the intensity of such signal to those obtained on the plateaux of constant thickness, where well known amounts of CoO have been deposited as measured by a quartz microbalance. For the regions shown in Fig. 1 the value varies in the range 1 nm $\leq t_{CoO} \leq$ 5 nm. Correspondingly, it is clearly seen that the shape and dimensions of the Fe magnetic domains dramatically change. In particular, at t_{CoO}=2 nm [panel (b)] a dense pattern of very small domains is observed, to which we will refer hereafter as "nanodomains." They are magnetized in the plane of the film, either parallel or antiparallel to one of the magnetic easy axes of Fe, which belong to the ⟨100⟩ family.

The nanodomains have been observed for 1.5 nm $\lesssim t_{CoO} \lesssim$ 3.8 nm, and also when decreasing the sample temperature, down to about 170 K. Below [panel (a)] and above [panels (c) and (d)] such thickness range the nanodomains gradually disappear and large FM domains take place. Such large domains have dimensions well above the microscope field of view and DW hundreds of nanometers wide, as expected in a conventional picture.[11,12] The observed micromagnetic structure clearly resembles that seen on top of Fe/NiO/Fe trilayers.[12,13] Moreover, also the evolution of the FM domains morphology as a function of the AFM thickness holds several similarities with that of Fe/NiO/Fe trilayers prepared under analogous conditions.[13,17] In particular, nanodomains are seen in a narrow AFM thickness range in both cases, while they coalesce to larger domains, similar to those of the substrate, for other thicknesses.[13]

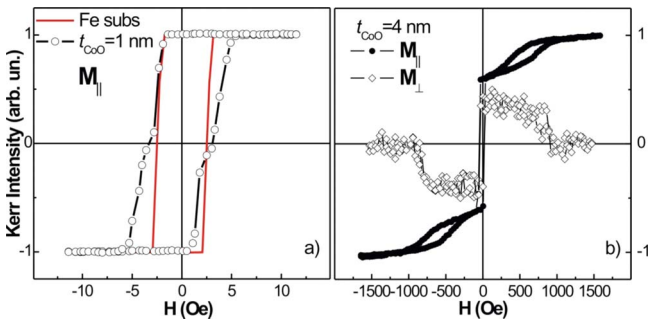

FIG. 2. (Color online) Panel (a): $\mathbf{M}_{\|}$ hysteresis loops from the Fe substrate (red line) and from a Fe/CoO/Fe trilayer with t_{CoO} =1 nm (open dots). Panel (b): $\mathbf{M}_{\|}$ (full dots) and $\mathbf{M}_{\perp}$ (open diamonds) hysteresis loops from a Fe/CoO/Fe trilayer with t_{CoO} =4 nm. All loops have been taken at T=200 K. The upper Fe layer has a constant thickness of 7 nm.

One of the most interesting features of the Fe/NiO/Fe case is that the NiO thickness (t_{NiO}) region corresponding to the nanodomains structure is in the same range of a critical thickness t_c between two different magnetic coupling regimes. The latter were characterized by either 90° in-plane ($t_{NiO} < t_c$) or parallel ($t_{NiO} > t_c$) alignment between the magnetization vectors in the two Fe films,[8] with t_c typically between 3 and 5 nm.[13] In order to study similar phenomena on the CoO based trilayers, magneto-optical Kerr effect (MOKE) has been employed for measuring magnetization hysteresis loops. The MOKE measurements give information, respectively, on the in-plane parallel ($\mathbf{M}_{\|}$) and in-plane perpendicular ($\mathbf{M}_{\perp}$) components of the layers' magnetization with respect to the external field $\mathbf{H}$. The latter was applied along one of the easy axes of Fe.[8] Details of the MOKE experimental setup are reported in Ref. 22.

Figure 2 reports some MOKE hysteresis loops taken on our Fe/CoO/Fe trilayers. We observe two radically different magnetization couplings between the Fe layers for either low or high t_{CoO}. Panel (a) displays MOKE hysteresis loops of the $\mathbf{M}_{\|}$ component related to the Fe substrate (red line) and to a Fe/CoO/Fe trilayer with t_{CoO}=1 nm (open dots) both taken at T=200 K. The former is a sharp square loop, with coercive field below 5 Oe, as already observed in similar films.[8] The latter loop testifies that the coupling between the Fe magnetization directions in the Fe/CoO/Fe structure is parallel, while the steps that occur during the reversal are likely due to unequal coercive fields for the Fe layers, on account of their quite different thickness.[8] Qualitatively similar loops have been observed for trilayers with 1 nm $\lesssim t_{CoO} \lesssim$ 2 nm. No signal from MOKE measurements of the $\mathbf{M}_{\perp}$ component was detected in these low t_{CoO} cases supporting the parallel coupling interpretation. Figure 2(b) shows hysteresis loops taken on a Fe/CoO/Fe trilayer with t_{CoO}=4 nm at T =200 K for both $\mathbf{M}_{\|}$ (full dots) and $\mathbf{M}_{\perp}$ (open diamonds) components. The $\mathbf{M}_{\|}$ loop can be considered as a superposition of two contributions. The first one is a central narrow loop attributed to the thick bottom Fe layer as confirmed by

comparison with the Fe substrate loop shown in Fig. 2(a). The outer loops occur at significantly higher magnetic fields (up to about 1000 Oe) and can be attributed to the thin upper Fe layer. In such a layer, the remanent magnetization direction is in the plane of the film (as also confirmed by X-PEEM) and at 90° with respect to that of the bottom layer. This is testified by the observation of the $\mathbf{M}_\perp$ component, also shown in Fig. 2, which is different from zero for $\mathbf{H}$ lower than the saturation field for the upper Fe layer (roughly 1000 Oe). The $\mathbf{M}_\perp$ loop has been arbitrarily rescaled on the vertical axis in order to be better displayed in the figure. Note that the IEC observed in our Fe/CoO/Fe trilayers has a dependence on the thickness of the AFM spacer which is opposite to that reported for Fe/NiO/Fe systems.[8] Even though the two systems are characterized by an analogous crystallographic quality (as measured by low-energy electron diffraction) and similar chemical interactions at the interfaces,[11,17] it must be underlined that several parameters may determine the relative direction among the magnetic moments at each FM/AFM interface including magnetic roughness, defects, and strain,[5,23] whose analysis is beyond the scope of this work. The IEC is in fact dependent on the mechanisms acting both on the individual interfaces and on the whole AFM spin structure. The modification of the IEC from parallel to in-plane perpendicular, observed when increasing t_{CoO}, is a clear evidence of the passing through a magnetic instability for a critical t_{CoO} in the range from 1.5 to 4 nm (considering the results of both X-PEEM and MOKE). This instability is evidently connected with the presence of nanodomains which survive only close to such critical thickness.

Strong variations have been reported for the characteristic temperatures of thin films of CoO coupled to magnetic layers, with respect to bulk CoO crystals, when decreasing t_{CoO}. In particular, the Néel temperature of CoO, which is relatively low (289 K) for bulk crystals, strongly increases with decreasing t_{CoO},[24] while the blocking temperature, i.e., the temperature above which exchange bias vanishes,[10] can become as low as 50 K for Fe_3O_4/CoO exchange biased structures with $t_{CoO} \simeq 1$ nm.[24] This could be a problem for the correct explanation of the Fe interlayer coupling, in particular for very thin CoO spacers (below 2 nm). For this reason, hysteresis loops have been acquired at different temperatures between 30 and 350 K. In this range, however, no significant differences have been seen on the loops, apart from an increase of the coercive fields, when decreasing the temperature. This observation suggests that the occurrence of a magnetic instability for 1.5 nm $\lesssim t_{CoO} \lesssim$ 4 nm is not influenced by the temperature in the range 30 K $< T <$ 350 K.

A deeper insight about the FM domain structure in the top Fe layer, at t_{CoO} close to the critical thickness, is given by the following analysis. As explained in Ref. 12, we expect for nanodomains a critical radius r_{min} below which they would collapse under the pressure exerted by the surrounding DW. The value of such r_{min} depends on the balance between the energetic cost associated with the DW surrounding the domain and the maximum energetic gain associated with the exchange coupling at the FM/AFM interface. The critical radius is thus a significant parameter to evaluate the strength of such a coupling. We estimate the value of r_{min} by measuring the minimum DW curvature radius from our images. This is done by first evaluating, from the measured pattern, the area A versus perimeter P distribution of the nanodomains. In Fig. 3 we have plotted such relationship for the nanodomains in Fig. 1(b).[25] We then consider that the A/P ratio is maximum for perfect circles. Such maximum ratio has been represented by plotting an additional line of equation $A = P^2/4\pi$ in Fig. 3. The intersection of the circles line with the power law best fitting the distribution is our experimental estimate of the minimum DW curvature corresponding to r_{min}. The few domains characterized by an A/P ratio higher than that of perfect circles would in fact correspond to unphysical artifacts due to noise and/or to the limited instrument resolution. From the plot shown in Fig. 3, we obtain $r_{min} \simeq 30$ nm for our Fe/CoO/Fe trilayers, very close to the value of 40 nm reported for Fe/NiO/Fe.[12] This indicates that the small magnetic domains imaged on the top layer have similar minimum sizes, driven by an interfacial exchange coupling of similar strength. Such a coupling is expected to fluctuate as a function of position giving rise to magnetic frustrations. A statistical analysis of the micromagnetic structure in the top Fe layer of our Fe/CoO/Fe trilayers reveals that the nanodomains are fractal, with fractal dimension of about 1.6, which is very similar to the value calculated for nanodomains on top of Fe/NiO/Fe systems.[12]

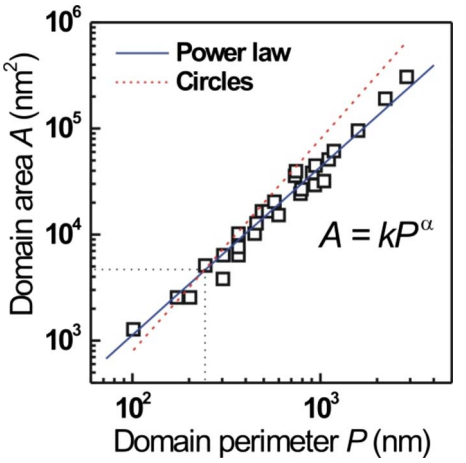

FIG. 3. (Color online) Area A versus perimeter P distribution of domain sizes as measured from the image in Fig. 1(b) (open squares). The continuous blue line is the linear best fit of the data; the dotted red line represents the A/P relation for perfect circles.

In conclusion, we have provided experimental evidences of the stabilization of very small in-plane magnetic domains on top of Fe/CoO/Fe trilayer systems and compared such observations with analogous results previously obtained on Fe/NiO/Fe systems, also in consideration of the dramatically different values of the magnetocrystalline anisotropy reported for the two systems. The observed domain structure cannot be explained in the framework of conventional magnetic domain theories, while it is clearly connected with magnetic instabilities generated by frustrations at the FM/AFM interfaces. Such instabilities are also expected to be connected to the different coupling regimes between the magnetization directions in the FM layers which have in fact been observed by MOKE for different CoO thicknesses.

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