

Perpendicular Interlayer Coupling in Ni₈₀Fe₂₀/NiO/Co Trilayers

J. Camarero,* Y. Pennec, J. Vogel, M. Bonfim, and S. Pizzini
Laboratoire Louis Néel, CNRS, BP166, 38042 Grenoble Cedex, France

F. Ernult, F. Fettaf, F. Garcia, F. Lançon, L. Billard, and B. Dieny
CEA/Grenoble, DRFMC/SPINTEC and S2PM, 38054 Grenoble Cedex, France

A. Tagliaferri and N. B. Brookes
European Synchrotron Radiation Facility (ESRF), 38043 Grenoble Cedex, France
(Received 19 June 2002; published 7 July 2003)

An in-plane perpendicular magnetic coupling between Ni₈₀Fe₂₀ and Co has been found in NiFe/NiO/Co trilayers for a NiO thickness ranging from 4 to 25 nm by magneto-optical Kerr effect and x-ray magnetic circular dichroism measurements. In the easy magnetization direction of the Co layer, the Co coercive field H_C increases when the thickness of the NiO layer t_{NiO} increases. Because of the coupling, H_C is always larger than for NiO/Co bilayers with the same thicknesses. The saturation field of the NiFe layer H_S decreases when t_{NiO} increases, indicating a weakening of the coupling. Numerical simulations show that the presence of interface roughness combined with a small value of the NiO anisotropy can explain the observed 90° coupling.

DOI: 10.1103/PhysRevLett.91.027201

PACS numbers: 75.25.+z, 75.30.Et, 75.50.Ee, 78.70.Dm

The coupling between ferromagnetic layers separated by a nonferromagnetic layer has attracted a lot of interest during the past decades [1], both for fundamental reasons and because of applications in the development of magnetic read-heads and sensors. In metallic systems, exchange interactions are propagated by itinerant electrons and thus can be transmitted over relatively long distances. This (oscillatory) interaction is well understood with a Rudermann-Kittel-Kasuya-Yosida-type coupling modified by the discreteness of the spacer material [2]. For insulating spacers, the exponential decreasing interaction can be attributed to electron tunneling in the limit of ultrathin spacer layers [3]. In several systems, a noncollinear alignment of the magnetization directions of the two ferromagnetic (F) layers was found, which could be reproduced phenomenologically introducing a biquadratic coupling term in the energy equation of the system. Several models have been proposed to explain this biquadratic term [4], but it is clear that in the case of antiferromagnetic (AF) spacers the interlayer exchange coupling with the F layers as well as the spin structure of the AF must be considered [5,6]. For instance, a spiraling spin structure in the AF can lead to different angles between the magnetization axes of the two F layers.

Recent experimental studies have found a 90° interlayer exchange coupling in F/AF/F trilayers using Mn [7] and NiO [8] as spacer layers. Other studies, with FeMn as AF, have shown that the angle between the magnetization directions of the two F layers depends on the AF thickness [9,10]. In all these studies, identical top and bottom ferromagnetic layers (CoFe [7], Fe₃O₄ [8], Fe [9]) were used which complicated the separation of the magnetization contributions of the two F layers to the $M(H)$

curves. A difference in the intrinsic magnetic anisotropy of the two F layers (NiFe and Co) was used by the authors of Ref. [10] to indirectly observe a spiraling spin structure within the AF layer after a special field-cooling procedure, with an antiparallel alignment of the magnetization axes of the two F layers.

In this Letter we present the first direct experimental observation of a room temperature in-plane 90° coupling in F/AF/F trilayers by means of magneto-optical Kerr effect (MOKE) and x-ray magnetic circular dichroism (XMCD). Numerical simulations confirm that the randomness of the interfacial coupling at the AF/F interface as well as the small effective anisotropy in the AF are the keys to understanding this type of coupling.

We used two different F materials, Ni₈₀Fe₂₀ and Co, to exploit the large difference in the magnetic anisotropy. The low magnetocrystalline anisotropy of the NiO makes it a good AF candidate to extend the thickness in which the 90° coupling is present. The trilayers were grown in the same run at room temperature in zero field on corning glass substrates in a multisource sputtering chamber [11]. A buffer layer of 5 nm Cu was used to promote [111] texture and to avoid a three-dimensional growth mode of permalloy on the amorphous substrate [12]. The NiO layers were deposited at oblique incidence by rf sputtering from a NiO target whereas the Co and NiFe layers were deposited at normal incidence by dc sputtering. Finally, the samples were capped by a 3 nm Cu layer to prevent oxidation. The thickness of the NiO layer t_{NiO} ranged from 4 to 25 nm, whereas the thickness was equal to 10 and 2 nm for permalloy and Co, respectively. No in-plane magnetic anisotropy is observed for the as-deposited permalloy layer on Cu, while the oblique

incidence during NiO deposition is known to induce an in-plane uniaxial anisotropy in the subsequently deposited Co layer, with the easy-axis of magnetization in the direction perpendicular to the plane of incidence of the sputtered NiO [11,13]. MOKE and XMCD hysteresis loops were measured with the field applied in the plane of the layers, either parallel or perpendicular to the easy-axis of the Co layer, to characterize the magnetic behavior as a function of the NiO thickness in the easy- and hard-axis directions of the Co layer.

The combination of p -polarized light in longitudinal Kerr experiments and the simultaneous detection of the two orthogonal components of the reflected light allows the in-plane magnetization components of the samples parallel (M_{\parallel}) and perpendicular (M_{\perp}) to the field direction to be determined simultaneously. The difference of the two reflected light components is proportional to M_{\parallel} , whereas the sum (i.e., the total reflectivity) is related to M_{\perp} . No exchange bias has been detected at room temperature, as expected in low-anisotropy NiO-exchange coupled systems [13]. Typical hysteresis loops of our trilayers are displayed in Fig. 1, in this case for $t_{\text{NiO}} = 8$ nm and with the field applied parallel to the Co easy-axis. M_{\parallel} shows two magnetic transitions, a reversible one between -50 and $+50$ Oe (minor loops) and an irreversible one at $H_C \pm 70$ Oe (major loop). The M_{\perp} loop indicates that the reversible transition in M_{\parallel} corresponds to magnetization along a hard-axis, with a reversal proceeding by coherent rotation. This behavior strongly suggests that the permalloy layer is coupled perpendicularly to the Co layer through the NiO. This is confirmed by the MOKE $M(H)$ loops for a field applied along the Co hard-axis (not shown). Also in that case two transitions are observed in the M_{\parallel} loops, but the irreversible one is at lower fields than the reversible transition, suggesting

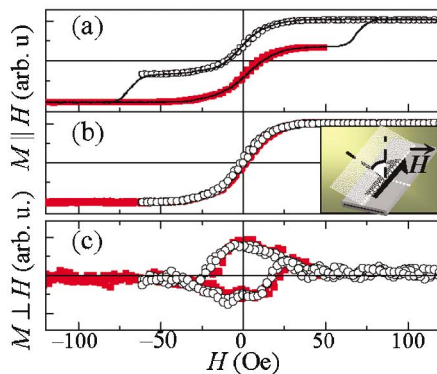


FIG. 1 (color online). Room temperature MOKE magnetization curves $M(H)$ of a 10 nm NiFe/8 nm NiO/2 nm Co trilayer film with H parallel to the Co easy axis. (a) M_{\parallel} major (solid line) and minor loops (symbols), (b) M_{\parallel} minor loops, (c) M_{\perp} . The inset shows the overall geometry for the growth process. The NiO plane of incidence (gray plane) is 55° off with respect to the normal sample surface (dark gray). The arrow indicates the field direction in the present experiment.

027201-2

that the hard-axis of Co corresponds to an easy-axis for $\text{Ni}_{80}\text{Fe}_{20}$.

In order to confirm this perpendicular coupling, element selective hysteresis loops were taken using XMCD. The experiments were carried out on the soft x-ray beam line ID8 of the European Synchrotron Radiation Facility. Chemical selectivity is obtained by tuning the photon energy to an absorption edge of the layer of interest [14]. In our case, maximum sensitivity was reached using the L_3 white lines of Co (778 eV) and Ni (853 eV) for the cobalt and NiFe layers, respectively. The x-ray absorption is measured by detecting the fluorescence yield, which allows measurements in the presence of a magnetic field and guarantees a large probing depth. The samples were mounted in a high vacuum chamber and the magnetic field was generated by an electromagnet mounted outside. The x rays were incident at 60° from the surface normal. The difference between the two field scans taken with the two opposite helicities of the x-ray beam gives the field dependence of the L_3 -XMCD intensity, and therefore the magnetization curve of the probed layer.

Figure 2 shows the field evolution of the L_3 -XMCD intensities of both Co and NiFe layers for the $t_{\text{NiO}} = 8$ nm trilayer with the applied field aligned parallel and perpendicular to the Co easy-axis. In the former case [Fig. 2(a)], the NiFe curve shows a fully reversible magnetization transition whereas an irreversible transition is observed for the Co layer. With the field applied perpendicular to the Co easy-axis, an almost perfect reversible transition is observed for the Co, while the NiFe shows an irreversible transition [Fig. 2(b)]. Therefore, XMCD confirms the perpendicular coupling between NiFe and Co layers across a NiO layer observed (indirectly) by MOKE.

MOKE hysteresis curves similar to those in Fig. 1 have been obtained for all NiO thicknesses t_{NiO} (up to 25 nm). Values of the Co coercive field H_C and the saturation field H_S of the NiFe layer are given in Fig. 3. For all t_{NiO} , we observe an enhancement of H_C with respect to NiO/Co

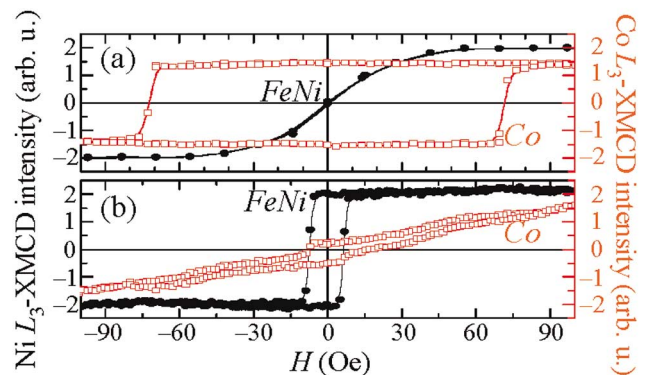


FIG. 2 (color online). (a) Layer selective hysteresis curves obtained with XMCD for Co (open circles, right-hand y axis) and NiFe (filled squares, left-hand y axis) with the field applied parallel (a) and perpendicular (b) to the Co easy-axis for a 10 nm NiFe/8 nm NiO/2 nm Co trilayer.

027201-2

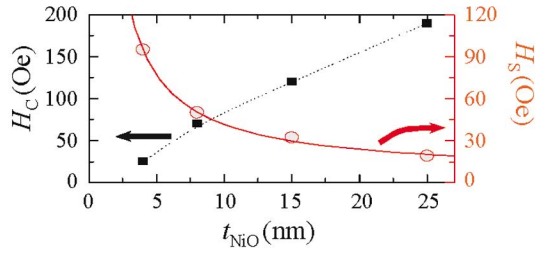


FIG. 3 (color online). NiO thickness dependence of the Co coercive field H_C (circles, left-hand y axis) and NiFe saturation field H_S (squares, right-hand y axis). The data are taken from $M(H)$ curves with the field applied parallel to the Co easy-axis. The dotted line is a guide for the eyes in $H_C(H)$ whereas the solid line represents a fit to the $H_S(H)$ data with the inverse of the NiO thickness (see text).

bilayers with the same thicknesses. As for the bilayers, the absence of exchange bias in the system is expected when the NiO magnetization is dragged by the reversal of the Co layer, as a result of the very small NiO grain size (~ 6 nm) in these samples [11,12]. We think that the increase of the coercivity with respect to NiO/Co bilayers is caused by the presence of the NiFe layer on the other side. Indeed, at each ferro/antiferro interface, frustration due to competing positive and negative exchange interactions takes place. This frustration leads within the antiferromagnet to the formation of an interfacial region of disordered magnetization in which magnetic relaxation can easily occur. The fact of having frustration at both interfaces in FeNi/NiO/Co therefore leads to more dissipation during the hysteresis loop and therefore larger coercivity than in NiO/Co bilayers. H_C also increases when the thickness of the NiO layer increases, as for NiO/Co bilayers [13]. Furthermore, H_S decreases inversely with t_{NiO} , indicating a decrease of the interlayer coupling. The energy per unit surface necessary to saturate the NiFe layer is related to the energy of a 90° domain wall E_{DW} in the NiO layer. In analogy with the Bloch wall energy in ferromagnetic systems, for an AF spacer of N layers, E_{DW} can be written as

$$E_{\text{DW}} \approx J_{\text{AF}} \sum_{i=1}^N 1 - \cos(\Theta_{i+1} - \Theta_i), \quad (1)$$

where J_{AF} is the exchange energy in the AF layer and Θ_i represents the angle of the magnetization in the i layer with respect to the AF anisotropy direction. Supposing a 90° domain wall, $\Theta_{i+1} - \Theta_i = \frac{\pi/2}{N}$, the extended sum can be approximated by $\pi^2/8N$. The domain wall energy can hence be written as $E_{\text{DW}} \propto J_{\text{AF}} \pi^2/8N$ with $N = t_{\text{AF}}/d_{\text{AF}}$, where d_{AF} is the AF interlayer distance. Finally,

$$H_S M_S^{\text{FeNi}} t_{\text{FeNi}} \approx J_{\text{AF}} \frac{\pi^2 d_{\text{AF}}}{8 t_{\text{AF}}}, \quad (2)$$

where M_S^{FeNi} and t_{FeNi} are the magnetization saturation value (730 emu/cm^3) and thickness (10 nm) of the NiFe

layer, respectively. H_S is therefore inversely proportional to the thickness t_{AF} as observed experimentally. As an example, for $t_{\text{NiO}} = 5$ nm, the 90° domain wall energy calculated from the saturation field of the NiFe layer using (2) is equal to 0.12 erg/cm^2 .

In order to reach a better understanding of the perpendicular interlayer coupling, we performed numerical simulations to investigate the spin configuration of the system. The latter is described as a three-dimensional cubic lattice of Heisenberg spins, with a length of 50 and a width of 32 lattice points. For the thickness, a nominal number of layers of 13 was taken for $\text{Ni}_{80}\text{Fe}_{20}$, 14 for NiO and seven for Co, with a total of 34 layers. A randomly generated roughness was introduced at both F/AF interfaces. The total energy E of the trilayer contains three contributions: (i) the Zeeman energy for each layer, (ii) the exchange coupling inside the layers and between the layers at the interface, and (iii) the magnetic anisotropy K of each layer. A phenomenological coupling was introduced in the proximity magnetism model describing changes in the exchange coupling across the AF layer with thickness fluctuations [5]. An algorithm based on the conjugated-gradient technique was developed to minimize the energy of the whole trilayer for each value of the applied magnetic field. The exchange coupling constants were estimated from the Néel and Curie temperatures of the AF and F bulk materials, respectively. The anisotropy constants have been estimated from experiments. Within experimental accuracy, the NiFe layer was found to be isotropic so that $K_{\text{FeNi}} = 0$. The in-plane anisotropy of Co ($K_{\text{Co}} = 5 \times 10^5 \text{ erg/cm}^3$) was obtained from measurements on NiO/Co bilayers [13]. The same measurements were also used to estimate the effective anisotropy of the NiO. From the NiO thickness dependence of the Co coercivity, this anisotropy was estimated to be $K_{\text{NiO}} = 4 \times 10^4 \text{ erg/cm}^3$ [13]. We explain the weak value of the NiO anisotropy in our layers in terms of a coherent magnetoelastic effect due to the deposition at oblique incidence. The magnetocrystalline anisotropy of the NiO is smeared out by the polycrystalline nature of our films consisting of very small grains with random in-plane crystallographic directions. The same easy magnetization axes have been taken for the Co and NiO layers.

Though the model does not take into account all of the details of the real sample, such as the frustration inside the NiO layer (grain boundaries) and chemical intermixing at the interfaces, the qualitative results agree very well with the experiment. In particular, the simulations reproduce the 90° interlayer coupling observed experimentally for $K_{\text{NiO}} < K_{\text{Co}}$. Figure 4 shows a cross-section view of the magnetic configurations of the system for saturation and zero field. At high fields, the magnetization in both ferromagnetic layers is aligned along the applied field whereas domain walls parallel and perpendicular to the interface can be observed in the AF layer [Fig. 4(a)]. This magnetic disorder in the AF layer originates from the interfacial roughness. As the lateral correlation length

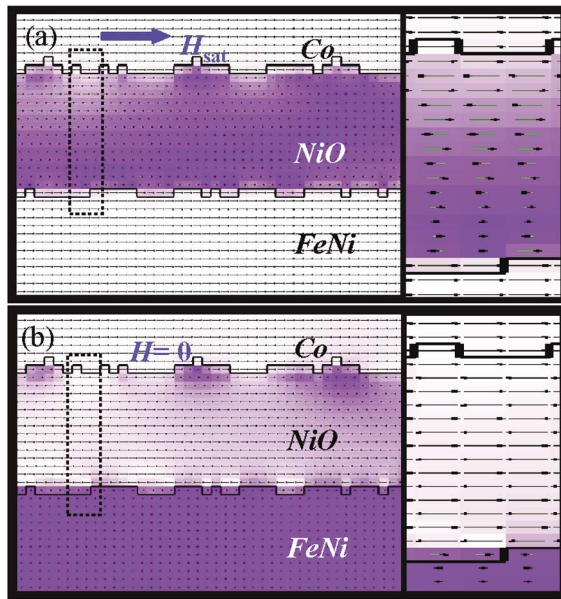


FIG. 4 (color online). Cross-section views of the simulated spin configurations of a FeNi/NiO/Co trilayer film at saturation (a) and zero field (b). The right-hand panels are magnetizations of the marked areas. The parameters in the simulation are $K_{\text{NiFe}} = 0$, $K_{\text{Co}} = 10 \times K_{\text{NiO}}$ (see text). The gray scale denotes the angle of the spins with respect to the field direction, from 0° (white) to $\pm 90^\circ$ (black). Note that both AF spin sublattices are shown.

of the interfacial roughness is smaller than the domain wall width, the AF layer cannot break into domains to locally satisfy the interfacial coupling. This results in an average perpendicular coupling between the ferromagnets and the spin lattice of the AF spacer, as in a spin flop configuration. Partial spirals in the NiO layer may be seen from one interface to the other [see Fig. 4(a), right-hand panel]. They do not, however, correspond to the homogeneous spiraling spin structure expected in Slonczewski's model [5]. This is due to the inclusion of the AF anisotropy in the energy of the system and from the local distortion of the magnetization introduced by the interfacial steps.

A completely different spin configuration is found at zero field [Fig. 4(b)]. Since there is no magnetic field to align the magnetization of the F layers, the spins of the AF spacer rotate in order to minimize their anisotropy energy. In this configuration the Co and NiO spin lattices are aligned in the same direction. The unwinding of the partial spiraling spin structure in the AF drags the magnetization of the adjacent NiFe layer, while the magnetization of the Co layer remains aligned along its easy-axis of magnetization. This leads to a 90° net angle between the magnetic moments of the two F layers.

In x-ray photoelectron emission microscopy experiments, a perfectly parallel alignment was observed at zero field between the interface spins of a NiO(100) single crystal and those of Co or Fe thin films deposited on top [15]. The apparent disagreement with our finding of a

perpendicular alignment at the NiFe/NiO interface may be explained by the negligible NiFe anisotropy and a higher interface roughness in our sputtered layers.

A crucial ingredient for the observation of the 90° interlayer coupling in the simulations is the condition $K_{\text{Co}} > K_{\text{NiO}} > K_{\text{NiFe}}$. For $K_{\text{Co}} = K_{\text{FeNi}} > K_{\text{NiO}}$, for example, the Co and NiFe moments are parallel, while the NiO moments are at 90° (spin flop).

In summary, we have presented direct evidence of perpendicular magnetic interlayer coupling in NiFe/NiO/Co trilayers for NiO thickness up to 25 nm. The strength of this coupling decreases inversely with t_{NiO} , as a result of the formation of spiraling spin structures in the NiO layer going from one F/AF interface to the other. Numerical simulations show that the presence of interfacial roughness combined with a small value of the effective NiO anisotropy can explain the observed 90° coupling. This knowledge opens new possibilities for atomic-scale engineering of magnetic properties. By fine-tuning the thickness and the anisotropy of both AF and F layers, it should be possible to control with great accuracy both the magnetic hardness and the interlayer coupling strength of any given F/AF/F trilayer system.

J.C. acknowledges the support of the European Community through Contract No. HPMF-CT-1999-00151.

*Present address: Departamento Física de la Materia Condensada, Universidad Autónoma de Madrid, 28049 Madrid, Spain.

Electronic address: julio.camarero@uam.es.

- [1] See, for instance, A. Fert and P. Bruno, in *Ultrathin Magnetic Structures II*, edited by B. Heinrich and J. A. C. Bland (Springer-Verlag, Berlin, 1994), p. 82; H. Hathaway, *ibid.*, p. 45.
- [2] P. Bruno and C. Chappert, *Phys. Rev. Lett.* **67**, 1602 (1991); *Phys. Rev. B* **46**, 261 (1992).
- [3] J. C. Slonczewski, *Phys. Rev. B* **39**, 6995 (1989); P. Bruno, *Phys. Rev. B* **49**, 13 231 (1994).
- [4] See, for instance, S. O. Demokritov, *J. Phys. D* **31**, 925 (1998), and references therein.
- [5] J. C. Slonczewski, *J. Magn. Magn. Mater.* **150**, 13 (1995).
- [6] H. Xi and R. M. White, *Phys. Rev. B* **62**, 3933 (2000).
- [7] M. E. Filipkowski *et al.*, *Phys. Rev. Lett.* **75**, 1847 (1995); V. Chakarian *et al.*, *Phys. Rev. B* **53**, 11 313 (1996).
- [8] P. A. A. van der Heijden *et al.*, *Phys. Rev. Lett.* **82**, 1020 (2000).
- [9] S.-S. Yan *et al.*, *Phys. Rev. B* **59**, R11 641 (1999)
- [10] F. H. Yang and C. L. Chien, *Phys. Rev. Lett.* **85**, 2597 (2000).
- [11] M. Cartier *et al.*, *J. Magn. Magn. Mater.* **223**, 63 (2001).
- [12] M. Cartier, Ph.D. thesis, Joseph Fourier University, 2000.
- [13] J. Camarero *et al.*, *Phys. Rev. B* **64**, 172402 (2001); *J. Appl. Phys.* **89**, 6585 (2001).
- [14] C. T. Chen *et al.*, *Phys. Rev. B* **48**, 642 (1993).
- [15] H. Ohldag *et al.*, *Phys. Rev. Lett.* **86**, 2878 (2001).