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Oscillatory interlayer exchange coupling in MgO tunnel junctions with perpendicular magnetic anisotropy

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The influence of magnetic layer thickness on the interlayer coupling through a tunnel barrier is investigated in Co/MgO/Co structures with perpendicular anisotropy. Despite the rather large MgO thickness, a clear antiferromagnetic coupling is observed. It oscillates with increasing magnetic layer thickness, in agreement with theories on indirect coupling through insulating spacers. Although the average interlayer coupling strength decreases with increasing annealing temperature, constant period and amplitude of these oscillations are observed for all annealing temperatures.

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The discoveries of antiferromagnetic (AF) exchange coupling between Fe layers through a Cr spacer[1] and of giant magnetoresistance in magnetic multilayered structures[2] opened the way to important technological developments in the field of spintronics.[3–5] These discoveries triggered numerous experimental and theoretical investigations.[6] In particular, the oscillatory character of exchange coupling as a function of the spacer and ferromagnetic (FM) layer thicknesses in metallic sandwiched structures was observed.[7–9]

Recently, magnetic tunnel junctions (MTJs) with MgO as an insulating spacer has become an object of a high interest for the spintronics community due to prediction[10,11] and observation[12,13] of extremely high tunnel magnetoresistance (TMR) ratios. In addition, indirect AF coupling across an insulator has been observed in fully epitaxial Fe/MgO/Fe MTJs structures,[14,15] as well as in Fe/MgO/γ-Fe_2O_3 (Ref. 16) and Fe_3O_4/MgO/Fe_3O_4 (Ref. 17) ones. This AF coupling strongly increases in amplitude as the MgO thickness is reduced down to a critical thickness below which pinholes formation takes place, leading to direct ferromagnetic coupling between magnetic electrodes. AF coupling agrees with existing theories,[18,19] and has been ascribed, through *ab initio* calculations, to either oxygen vacancies in the MgO barrier[15] or oxidation of the magnetic electrodes.[17]

On another hand, there is a growing interest in magnetic tunnel junctions with perpendicular to the FM/spacer interface magnetized electrodes (p-MTJs) originating from perpendicular magnetic anisotropy (PMA). These structures are known for their larger thermal stability and greater efficiency for spin transfer torque switching.[20] However, there exist very few basic studies on the indirect coupling in these structures. Using an insulating NiO spacer separating two (Co/Pt) multilayers, Liu and Adenwalla[21] observed an increasing indirect coupling as a function of measurement temperature, as predicted by Bruno.[19] At the same time, they observed, contrary to the above-mentioned theories, an oscillatory variation in the coupling with spacer thickness, which they tentatively attributed to the antiferromagnetic nature of the insulating NiO spacer.

In MgO-based magnetic tunnel junctions with perpendicular magnetic anisotropy prepared by a sputtering technique, we recently evidenced the existence of AF coupling between magnetic electrodes, and studied its variation with barrier thickness, as well as with annealing (T_A) and measurement temperatures.[22,23] AF coupling strength increases with increasing measurement temperature, in agreement with Bruno's theory.[19] It also appears to be strongly dependent on the perpendicular magnetic anisotropy of the structures studied, being larger for smaller PMA values. It has been recently predicted that roughness induced dipolar coupling (Néel coupling[24]) in perpendicularly magnetized structures could turn from ferromagnetic to antiferromagnetic when increasing PMA.[25] However, coupling strength should in this case increase with decreasing measurement temperature or increasing PMA (increasing T_A), contrary to our observations.

We present here a systematic study of AF coupling dependence on the magnetic electrode thickness in MgO-based p-MTJs. In the investigated samples, PMA is mainly of an interfacial origin so that its amplitude per unit volume decreases monotonically with increasing magnetic layer thickness. At the same time, AF coupling shows an oscillatory behavior with a period of about 0.8 nm for FM layer thicknesses up to 3 nm. Moreover, these oscillations persist with essentially the same amplitude and period for annealing temperatures up to 350 °C while the average AF coupling energy tends to decrease with increasing T_A.

Samples were prepared by dc sputtering onto thermally oxidized (500 nm SiO_2) Si (100) oriented substrates at ambient temperature, with typical deposition rates of about 0.05 nm/s. Multilayer structures consists of $Si/SiO_2\|Ta_3/Pt_{20}/Co_{1.2}/MgO_{1.3}/Co_t/Pt_3$ with indices representing thickness (in nanometer). For the top Co electrode, t varies from 1.0 to 3.2 nm. More than 20 different samples were prepared in a random order over a 1 year interval. The MgO barrier is obtained through two successive deposition/oxidation steps of a metallic Mg layer 0.7 nm thick, oxidation being carried out for 10 min under 900 mbar oxygen pressure. The actual barrier thickness (1.3 nm) is estimated to account for a 20% isotropic volume decrease in the atomic cell upon oxidation. Samples were annealed in vacuum for 30 min from 250 up to 350 °C.

The choice of Co as FM electrodes and natural oxidation of the Mg layers leads to an optimized perpendicular magnetic anisotropy[23] and allows maximizing both FM layer thickness and annealing temperature ranges in which both electrodes are magnetized out of plane. The 1.2-nm-thick bottom Co layer grows on a 20-nm-thick Pt buffer. As a consequence, its magnetization is always out of plane and its anisotropy is larger than the one of the top Co layer for all annealing temperatures and top Co thickness ranges investigated.

On another hand, the lack of epitaxial relationship between pure Co layers [hexagonal (0001) or face-centered-cubic (111) planes] and MgO barrier [(100) oriented rock-salt structure] is not expected to lead to a high TMR amplitude. Indeed, a TMR ratio of only 10% is measured on $Co_{1.2}/MgO/Co_{1.2}$ structures after annealing at 350 °C.

In order to probe the out-of-plane component M_z of the magnetization, magnetic hysteresis loops were recorded on unpatterned samples by extraordinary Hall effect at room temperature in a classical Van der Pauw geometry. The magnetic field was applied perpendicular to the layer plane and calibrated using a Hall sensor. Zero value of the magnetic field was further checked by measuring the magnetoresistive response of a 50-nm-thick $Ni_{80}Fe_{20}$ (Permalloy) film. The sign and amplitude of the magnetic coupling between both magnetic electrodes were determined from minor hysteresis loops (with the magnetization of the softest top layer reversal only). All minor loops were recorded with the same frequency (0.2 Hz) and field step (5 Oe). Negative values of the interlayer coupling field (energy) correspond to a preferred antiparallel alignment of the magnetization directions on both sides of the MgO barrier. Vibrating sample magnetometry (VSM) and superconducting quantum interference device (SQUID) measurements were carried out to extract absolute values of the magnetizations of both top and bottom Co layers.

Figure 1 shows the Hall hysteresis loops of $Co_{1.2}/MgO_{1.3}/Co_t$ (nanometer) structures after annealing at 320 °C. Both major and minor hysteresis loops are shown in Fig. 1(a) for a top Co thickness of 1.2 nm. Coming from negative saturation, the transition of both soft (at 260 Oe) and hard (at 2400 Oe) layers are of equal amplitudes. This implies equal Hall coefficients for Pt/Co/MgO and MgO/Co/Pt stacks. In addition, the minor loop is shifted by −60 Oe [see Fig. 1(c)], indicating preferred antiferromagnetic coupling between magnetic electrodes. It must be noted that in previous studies of fully epitaxial structures with in-plane magnetizations,[14–17] the coupling always turned to be ferromagnetic for MgO thicknesses larger than 0.8–0.9 nm. In our case, AF coupling, although small, is observed through a much thicker 1.3 nm MgO layer.

Figures 1(b)–1(e) display minor hysteresis loops recorded for increasing top Co thickness. One can clearly observe an oscillation of the coupling field amplitude with increasing top Co layer thickness while its sign remains negative whatever Co thickness, indicating preferred antiferromagnetic coupling. The squareness of the minor loops (nucleation field/saturation field ratio) progressively decreases with increasing top Co thickness above 2.0 nm, as a consequence of the decreasing perpendicular anisotropy.

A more quantitative insight is given in Fig. 2, which shows the variation in the coupling field as a function of top Co thickness for increasing T_A. As a consequence of the strong PMA increase with increasing T_A,[23] the critical top Co thickness (transition from out-of-plane to in-plane configuration) increases from 1.4 nm for $T_A=250$ °C to more than 3.0 nm for $T_A=350$ °C. Interlayer coupling is essentially antiferromagnetic for all ranges of top Co layer thickness and annealing temperatures investigated. The most striking feature is that coupling keeps oscillating as a function of the top magnetic layer thickness up to at least 3 nm. Although the average coupling field strength progressively decreases with

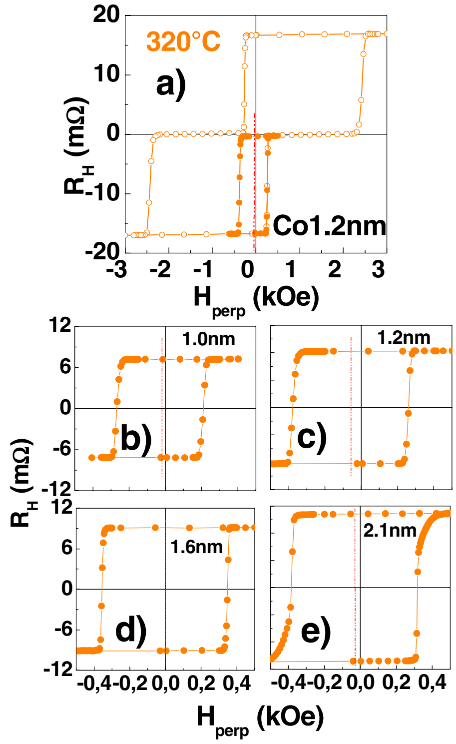

FIG. 1. (Color online) Hall hysteresis loops for $Co_{1.2}/MgO_{1.3}/Co_t$ structures after annealing at 320 °C. Magnetic field is applied perpendicular to the sample plane. Curve (a) shows both major and minor loops for $t=1.2$ nm. Curves (b)–(e) show minor loops for increasing top Co thickness. The vertical bar materializes the field shift of the minor loop.

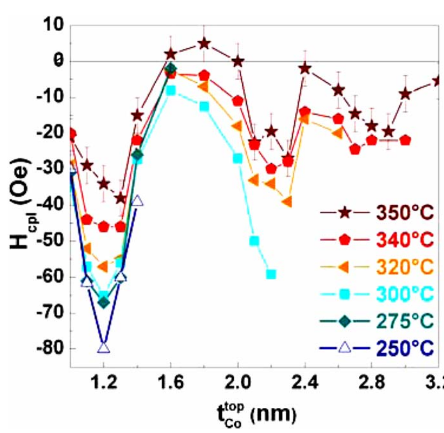

FIG. 2. (Color online) Variation in the indirect coupling field with top Co thickness for different annealing temperatures. Uncertainty on coupling field is taken equal to the field step (5 Oe) used in Hall measurements for minor hysteresis loops and is only represented for $T_A=350$ °C.

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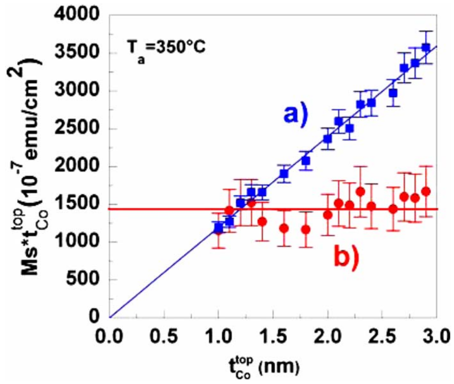

FIG. 3. (Color online) Variations as a function of top Co thickness, after annealing at 350 °C, of the magnetic moments of (a) top and (b) bottom electrodes. The blue line has a slope of 1200 emu/cm^3 and zero intercept, whereas the horizontal red line corresponds to $1200\times 1.2\times 10^{-7}$ emu/cm^2.

increasing T_A, period and amplitude of the oscillation are mostly independent of annealing.

The coupling energy is determined from the coupling field H_{cpl} using the relation,

$$J = H_{cpl}(M_{sT}tM_{sB}t_B)/(M_{sT}t + M_{sB}t_B), \qquad (1)$$

where t and t_B (1.2 nm) represent thicknesses of the top and bottom magnetic electrodes, respectively, and M_{sT} and M_{sB} are their saturation magnetizations.

Hall measurements cannot be used to extract absolute values of the magnetizations since Hall resistance depends on both magnetization and Hall coefficient of the magnetic layers, the latter increasing substantially with annealing treatments.[26] In order to extract values of magnetizations of both bottom and top electrodes, we carried out direct magnetic VSM and SQUID measurements. Our analysis relies upon the hypothesis that low-temperature saturation magnetization, for Co electrodes thicker than 1.0 nm, equals that of bulk cobalt, that is, 1430 emu/cm^3. We first carried out low-temperature SQUID measurements on a $Co_{1.2}/MgO/Co_{1.2}$ structure and observe a 16% reduction in magnetization with temperature increasing from 5 to 300 K. We thus deduce a saturation magnetization for the 1.2-nm-thick magnetic electrodes of 1200 emu/cm^3 at room temperature and this value is used to calibrate our VSM measurements. The corresponding magnetization, determined from VSM measurements on the whole series of samples after annealing at 350 °C, multiplied by the top magnetic thickness, is shown in Fig. 3(a) as a function of the top Co layer thickness. The expected linear variation with a slope of 1200 emu/cm^3 and zero intercept is superimposed to the data points. The good agreement with experimental data (6% scatter) shows that saturation magnetization is independent of Co thickness between 1 and 3 nm. The quite small scatter of M_{sT} values, which mainly comes from experimental resolution and accuracy in the determination of sample surface, implies a rather good stability of our sputtering unit.

The same procedure is used for the bottom Co electrode and the results (saturation magnetization multiplied by 1.2 nm) are shown in Fig. 3(b). They reasonably agree with the horizontal line corresponding to $1440(1200\times 1.2)10^{-7}$ emu/cm^2. The larger scatter (20%) observed in Fig. 3(b) compared with Fig. 3(a) is related to both larger applied field and larger field interval required to switch the magnetically harder bottom layer, making the determination of its amplitude more difficult.

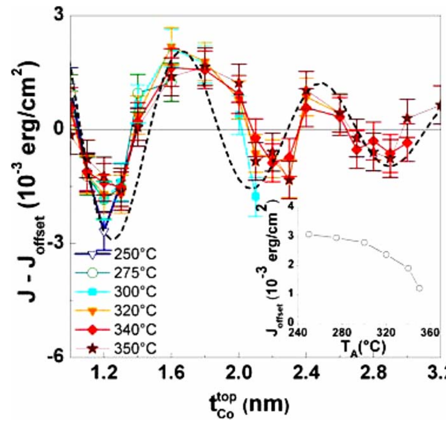

FIG. 4. (Color online) Variation in the coupling energy as a function of top Co thickness, calculated from data of Figs. 2 and 3 according to Eq. (1). Coupling energies, after allowing for a vertical offset $J_{offset}(T_A)$ given in the inset, are fitted to Eq. (2) (dotted black line).

Based on these results, we will now assume that saturation magnetization values obtained from Fig. 3 (1200 emu/cm^3) can be used for all annealing temperatures for the determination of coupling energies according to Eq. (1). Figure 4 gives the corresponding variation in coupling energy as a function of top Co thickness. In order to make easier the analysis of the coupling oscillations on the whole range of annealing temperatures, all curves are superimposed to oscillate around zero energy, the vertical energy offset $J_{offset}(T_A)$ [included as a fitting parameter to Eq. (2) shown below] being given in the inset.

Figure 4 demonstrates that curves for different annealing temperatures coincide, highlighting the constant oscillation amplitude and period for all T_A used. $J_{offset}(T_A)$ progressively decreases with increasing annealing temperature. Such a decrease in coupling strength has been observed in all MgO-based perpendicular junctions we studied up to now. It could be due to a progressive deoxidation of the bottom magnetic electrode with increasing annealing temperature. This mechanism seems to be in agreement with *ab initio* calculations,[17] which relate deoxidation of the magnetic electrodes to a decreasing AF coupling strength. This also confirms our previous experimental results concerning perpendicular anisotropy in $Pt/Co/AlO_x$ trilayers[26] in which the increase in the anisotropy with annealing for long oxidation times was also explained by a deoxidation of the magnetic layer followed by the formation of magnetic metal-oxygen bonds at the interface with the insulating barrier.

This oscillatory variation in coupling as a function of magnetic layer thickness, predicted by theory,[19] has been confirmed only in the case of a metallic spacer with in-plane magnetized electrodes.[9] The rather large oscillation amplitude we observe here is certainly related to the presence of the insulating MgO barrier. This effect has been recently evidenced in MgO-capped Fe/Cr/Fe structures.[27]

According to Bruno's theory,[19] the oscillatory variation in the coupling as a function of magnetic layer thickness originates from Fabry-Perot-type interferences of the electron Bloch wave multiple reflections in FM layers. We apply this model to present studies and fit the corresponding J data to the following expression valid in the approximation of large layer thicknesses:

$$J = A/C^2 \sin(2\pi t/\Lambda + \Phi) \tag{2}$$

with $C=1+(k_F t)/(k_F^{\downarrow} D)$, and where A is a constant and t and D (1.3 nm) represent the thickness of the top magnetic electrode and barrier, respectively. Λ and Φ indicate the oscillation period and phase while $k_F^{\downarrow}=\pi/\Lambda$ is the Fermi wave vector of the spin-down electrons in the ferromagnetic layer and k_F the imaginary part of the Fermi wave vector in the insulating spacer.

We should note here that despite the limitations of Bruno's model to the case of a transparent barrier, Eq. (2) gives a reasonable agreement with experiment for both period and damping of the oscillations (with $\Lambda=0.85$ nm, $\Phi=0.6\pi$, and $k_F=5\ \text{nm}^{-1}$). The maximum at about 1.7 nm is clearly broader than the other ones, suggesting the possibility of additional contributions with a different period.[9] However, the Co thickness range investigated in this study (which is limited by perpendicular anisotropy properties) does not allow us to try fitting our experimental data to more than one oscillation period.

The obtained k_F value of 5 nm^{-1} represents intermediate barrier quality between ideal epitaxial MgO and amorphous Al_2O_3 cases. The oscillation period of 0.85 nm corresponds to 4.2 monolayers (MLs) for (111) fcc and (0001) hcp Co or 4.9 ML for (001) fcc Co. However, this value for (001) fcc Co is different from that reported so far (3.5 ML) for (001) Co/Cu/Co structures.[9] This suggests that Co structure in our samples is closer to the case of (111) fcc crystal structure. This period leads to a wave vector $k_F^{\downarrow}$ of about 4 nm^{-1} which in this case should be the one for (111) fcc Co. Theoretical studies on the exchange coupling behavior in p-MTJs are in progress and will help to clarify this assumption.[28]

In conclusion, we have provided the evidence of AF interlayer exchange coupling oscillations as a function of Co layer thickness in MgO-based perpendicular MTJ structures prepared by dc sputtering. The AF coupling energy oscillations as a function of magnetic layer thickness are in agreement with theoretical predictions and compare well with similar experimental observations in the case of metallic spacers. Although average coupling strength progressively decreases with increasing annealing temperature, amplitude and period of the oscillations are found to be essentially independent of annealing temperature.

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[1] P. Grünberg, R. Schreiber, Y. Pang, M. B. Brodsky, and H. Sowers, Phys. Rev. Lett. **57**, 2442 (1986).
[2] M. N. Baibich, J.-M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988).
[3] B. Dieny, V. S. Speriosu, S. S. P. Parkin, B. A. Gurney, D. R. Wilhoit, and D. Mauri, Phys. Rev. B **43**, 1297 (1991).
[4] A. Fert, A. Barthélémy, J. Ben Youssef, J.-P. Contour, V. Cros, J. M. De Teresa, A. Hamzic, J.-M. George, G. Faini, J. Grollier, H. Jaffrès, H. Le Gall, F. Montaigne, F. Pailloux, and F. Petroff, Mater. Sci. Eng., B **84**, 1 (2001).
[5] S. A. Wolf, Science **294**, 1488 (2001).
[6] A. Fert and P. Bruno, in *Ultrathin Magnetic Structures*, edited by B. Heinrich and J. A. C. Bland (Springer-Verlag, Berlin, 1994), Vol. 2, Chap. 2.2, p. 82.
[7] S. S. P. Parkin, N. More, and K. P. Roche, Phys. Rev. Lett. **64**, 2304 (1990).
[8] S. S. P. Parkin, Phys. Rev. Lett. **67**, 3598 (1991).
[9] P. J. H. Bloemen, M. T. Johnson, M. T. H. van de Vorst, R. Coehoorn, J. J. de Vries, R. Jungblut, J. aan de Stegge, A. Reinders, and W. J. M. de Jonge, Phys. Rev. Lett. **72**, 764 (1994).
[10] W. H. Butler, X.-G. Zhang, T. C. Schulthess, and J. M. MacLaren, Phys. Rev. B **63**, 054416 (2001).
[11] J. Mathon and A. Umerski, Phys. Rev. B **63**, 220403(R) (2001).
[12] S. Yuasa, A. Fukushima, H. Kubota, Y. Suzuki, and K. Ando, Appl. Phys. Lett. **89**, 042505 (2006).
[13] S. Ikeda, J. Hayakawa, Y. Ashizawa, Y. M. Lee, K. Miura, H. Hasegawa, M. Tsunoda, F. Matsukura, and H. Ohno, Appl. Phys. Lett. **93**, 082508 (2008).
[14] J. Faure-Vincent, C. Tiusan, C. Bellouard, E. Popova, M. Hehn, F. Montaigne, and A. Schuhl, Phys. Rev. Lett. **89**, 107206 (2002).
[15] T. Katayama, S. Yuasa, J. Velev, M. Y. Zhuravlev, S. S. Jaswal, and E. Y. Tsymbal, Appl. Phys. Lett. **89**, 112503 (2006).
[16] H. Yanagihara, Y. Toyoda, and E. Kita, J. Appl. Phys. **101**, 09D101 (2007).
[17] H.-C. Wu, S. K. Arora, O. N. Mryasov, and I. V. Shvets, Appl. Phys. Lett. **92**, 182502 (2008).
[18] J. C. Slonczewski, J. Magn. Magn. Mater. **150**, 13 (1995).
[19] P. Bruno, Phys. Rev. B **52**, 411 (1995).
[20] S. Mangin, D. Ravelosona, J. A. Katine, M. J. Carey, B. D. Terris, and E. E. Fullerton, Nature Mater. **5**, 210 (2006).
[21] Z. Y. Liu and S. Adenwalla, Phys. Rev. Lett. **91**, 037207 (2003).
[22] L. E. Nistor, B. Rodmacq, S. Auffret, A. Schuhl, and B. Dieny, IEEE Trans. Magn. **45**, 3472 (2009).
[23] L. E. Nistor, B. Rodmacq, S. Auffret, and B. Dieny, Appl. Phys. Lett. **94**, 012512 (2009).
[24] L. Néel, C. R. Hebd. Séances Acad. Sci. **255**, 1676 (1962).
[25] J. Moritz, F. Garcia, J.-C. Toussaint, B. Dieny, and J.-P. Nozières, Europhys. Lett. **65**, 123 (2004).
[26] B. Rodmacq, A. Manchon, C. Ducruet, S. Auffret, and B. Dieny, Phys. Rev. B **79**, 024423 (2009).
[27] D. Halley, O. Bengone, S. Boukari, and W. Weber, Phys. Rev. Lett. **102**, 027201 (2009).
[28] H.-X. Yang, A. Kalitsov, and M. Chshiev (unpublished).