Short Period Magnetic Coupling Oscillations in Co/Si Multilayers: Theory versus Experiment

Nader Yaacoub, Christian Meny,* Olivier Bengone, and Pierre Panissod
Institut de Physique et Chimie des Matériaux de Strasbourg, UMR 7504 ULP-CNRS, 23 rue du Læss, BP 43, 67034 Strasbourg cedex 2, France
(Received 18 May 2006; published 22 December 2006)

Today the magnetic properties of multilayers and nanostructures including a metal or an insulator as a nonmagnetic spacer layer are rather well understood. But they are much more controversial for semiconductor spacers. For instance, for Co/Si multilayers short period coupling oscillations are predicted by *ab initio* computations but have yet to be observed. Here we show in Co/Si multilayers prepared at low temperature (90 K) strong saturation field oscillations that are consistent with the predicted coupling oscillations. However, the decay length of the oscillations is much longer than the expected one and cannot be explained within the framework of available theories.

DOI: 10.1103/PhysRevLett.97.257206 PACS numbers: 75.70.Cn, 68.35.Ct, 75.40.Mg

Since technological developments [1] allowed physicists to study the properties of thin films, multilayers, and nanostructures, many new properties have been evidenced [2–6]. However, in ferromagnetic-metal/semiconductor nanostructures the expected properties are altered by the large diffusion of metal and semiconductor atoms at the interfaces [7]. For example, for the thoroughly studied Fe/Si multilayer system, experimental results about the nature of the magnetic coupling (ferromagnetic and/or antiferromagnetic) between the Fe layers through the Si spacer are very controversial. Both oscillatory coupling [8] (when interfacial mixing is strong) and nonoscillatory antiferromagnetic (AF) coupling [9] (for sharper interfaces) are observed while theoretical calculations predict oscillatory coupling for perfect interfaces and nonoscillatory AF coupling when alloyed [10]. Although much less studied, similar discrepancies are observed when studying the properties of Co/Si multilayers [11–13]. In this Letter, we report the experimental observation of short period saturation field oscillations with the Si spacer thickness that are in agreement with the coupling oscillations predicted by *ab initio* calculations [11]. Such oscillations have never been observed experimentally up to now. This observation has been rendered possible by reducing the interfacial mixing by low temperature deposition (90 K) of the Co/Si multilayers. However, our experimental results show unusual features that were not predicted in the above calculations or by our own computations: up to 4 nm of spacer thickness the coupling strength does not decrease and the coupling strength is identical for low (10 K) and high (300 K) measurement temperatures. These results will renew the interest in ferromagnetic-metal/semiconductor systems and might lead to new technological developments in spin electronics.

The key point in metal/semiconductor systems is to avoid, as much as possible, mixing of the metallic and semiconductor atoms at the interfaces between the layers. Because this intermixing is likely to occur during the growth of the samples, we have chosen to prepare the samples at low temperature to limit the mobility of the atoms. To achieve this, the samples were prepared in an Alliance Concept sputtering system equipped with a nitrogen cooled sample holder. The sample holder temperature has been maintained at 90 ± 5 K during the whole sample preparation. The thermal contact between the sample holder and the samples was obtained by clamping the samples to the sample holder. The Si and Co layers are then sputtered in succession from pure targets at an identical sputtering rate of 0.05 nm/s. The resulting samples have the following architecture: Substrate Si⟨111⟩Si 5 nm/[Co 3 nm/Si t nm] $*$ 6/Si capping, the Si thickness "t" varying from 1 to 4 nm by 0.05 nm increments. The Si capping thickness is adjusted so that the topmost layer has a constant 10 nm thickness. Prior to the deposition, the Si substrates have been etched by argon plasma in order to remove the silicon native oxide at the surface of the samples.

Careful structural analyses of the samples have been performed. These analyses will be presented in details in another paper [14], but, since interface quality plays a crucial role in such a system, the main results about the interface quality are summarized here. Interface quality has been probed by zero field nuclear magnetic resonance (NMR) [15] and by low angle x-ray diffraction. Examples of NMR spectra are given in Fig. 1: increasing the Co thickness leads to an increase of the bulk Co contribution only. This shows that the intermixed region at the Co/Si interfaces is confined to a small thickness. The simulation of the x-ray and NMR data revealed an identical interface thickness for both techniques. The intermixed region at the Co/Si interface has been confined to 5 atomic planes. This interface thickness is much smaller than the one reported for samples prepared at room temperature (25 planes [7]). Such an interface thickness is similar to the one observed in metallic multilayers showing exchange coupling oscillations [15]. Finally, it must also be noted that while NMR

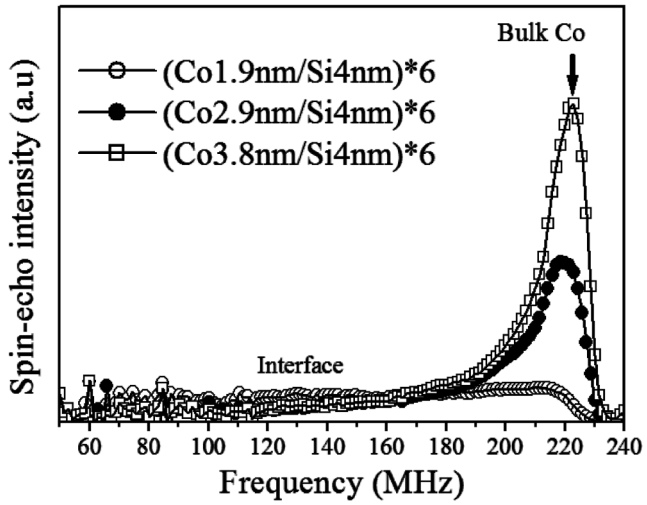

FIG. 1. NMR spectra of Co/Si multilayers. Increasing the Co thickness does increase the bulk contribution only. The interface width is limited by low temperature elaboration.

is sensitive to short range intermixing, x-ray diffraction is sensitive to both short and long range roughness. As both techniques give identical interfacial thickness, it shows that long range roughness is small in our samples and that the interfaces are composed by an intimate mixing of Co and Si atoms that is laterally homogenous on a very large scale. The flatness of the layers has been confirmed by atomic force measurements, showing an rms roughness as small as 2 Å at the top of the samples.

Magnetic properties of our samples are summarized in Fig. 2. Examples of magnetization curves, measured by a SQUID magnetometer at room temperature, are given in the top panel. As can be seen, saturation fields change dramatically within a few tenths of a nanometer. They vary typically between 500 to 5000 Oe. Because saturation fields are difficult to evaluate properly, we have plotted instead, in the bottom panel of Fig. 1, the energy needed to saturate the samples (integral of HdM) versus the Si thickness. Open symbols represent room temperature magnetization measurements, while solid dots show measurements performed at 10 K. Clear oscillations, with sharp maxima, are observed with an oscillation period of about 0.4 nm. Figure 2 also reveals two surprising features. First, for the Si thickness range under consideration, the magnitude of the oscillations does not vary significantly with the spacer thickness, and second, the magnitude of the oscillations is identical for both measurement temperatures. To illustrate this comment an example of magnetization curve obtained at 300 and 10 K for the sample with $t = 2.2$ nm is shown in Fig. 3. Apart from a small increase of coercive field the magnetization curve obtained at low temperature is identical to the one obtained at room temperature.

In the case of nonmetallic spacers, coupling oscillations are not usually expected [16]. Indeed, in the framework of the interference model and for semiconductor or insulator

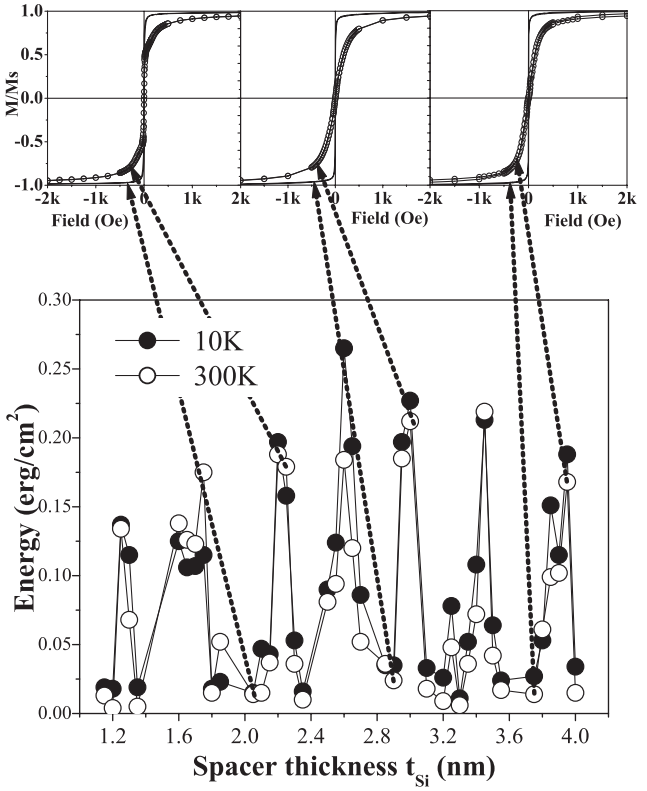

FIG. 2. Magnetic properties of the Co/Si multilayers versus the Si spacer thickness. Top panel: examples of magnetization curves. Bottom panel: magnetic energy needed to saturate the samples.

spacer layers, the magnetic coupling should be antiferromagnetic with a strength decaying exponentially with the spacer thickness [17]. However, *ab initio* computations performed by Enkovaara *et al.* suggest the presence of coupling oscillations in Co/Si multilayers. As observed

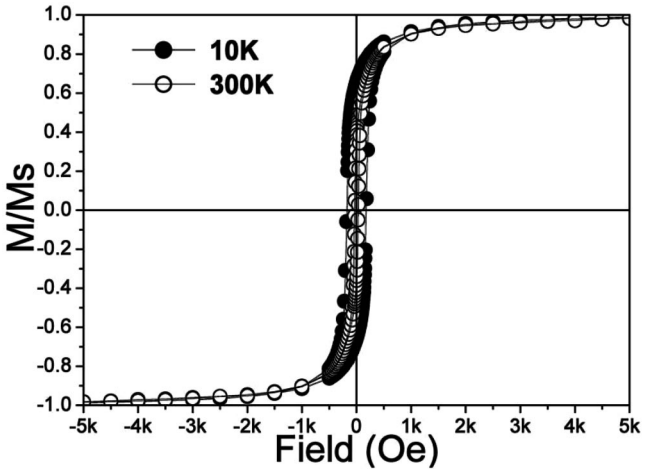

FIG. 3. Magnetization loops of a [Co(3 nm)/Si(2.2 nn)] * 6 multilayer. Temperature of measurements: 300 K (open symbols) and 10 K (solid symbols).

in our experimental work, these computations show that the sign of the magnetic coupling between the Co layers changes from ferromagnetic to antiferromagnetic each time one atomic layer of Si is added in the spacer. However, these computations were performed for multilayers composed by only two atomic planes of Co and with hexagonal silicon constrained to the lattice parameter of bulk Co. To confirm these results we have undertaken first-principles calculations for Co/Si/Co trilayers. In contrast with the previously cited work those computations were performed with fcc diamond silicon sandwiched between two infinite leads of fcc Co. The three layers where stacked along the ⟨100⟩ direction and the Si lattice was constrained by 7.5% to match the Co lattice parameter. The first-principles calculations are performed by means of the surface green function technique [18,19], based on the tight-binding linear muffin-tin orbitals method [20], within the atomic sphere approximation. We used the local spin density approximation in the Vosko-Wilk-Nusair parametrization scheme [21]. As shown in Fig. 4, even if the computation conditions are quite different from Enkovaara's work, ferromagnetic and antiferromagnetic ground states are also found to oscillate with a period of two Si atomic layers. It can be noted that the computed coupling strengths are more than 1 order of magnitude higher than the experimental ones. Even if the interfacial mixing in our samples is limited, one cannot rule out the possibility of some atomic scale Si thickness inhomogeneities leading to a distribution of coupling strengths. This will result, on one hand, in a reduction of the experimentally observed coupling strengths and, on the other hand, to a distribution of saturation fields as observed on the samples magnetization curve shapes showing rather high slopes close to zero field and high saturation fields.

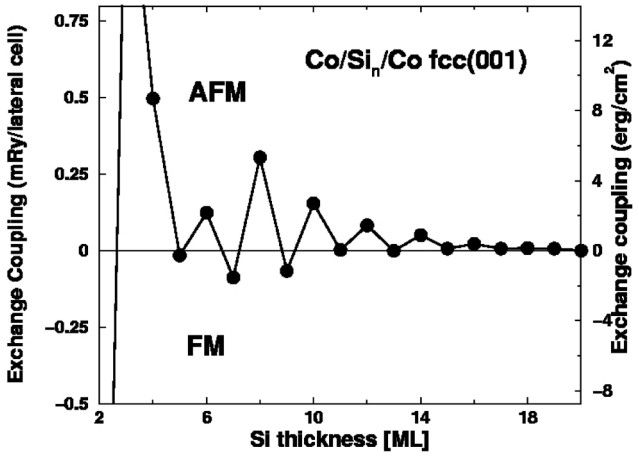

FIG. 4. Calculated exchange coupling of Co/Si(n)/Co multilayers as a function of silicon thickness. The coupling strength is given in mRy per computation shell (left scale) and in erg/cm^2 (right scale) for comparison with experimental values. Positive and negative values stand for antiferromagnetic (AFM) and ferromagnetic (FM) coupling, respectively, of the cobalt magnetic layers.

To understand the difference between the behavior predicted by the interference model and first-principles calculations, we must keep in mind that in the interference model [16] the coupling oscillations are computed from the shape of the bulk Fermi surface (real or complex) of the spacer layer. Therefore, no modification of the electronic structure of the spacer due to the finite size of the spacer layer is taken into account. In the case when one of the components of the multilayered system is a semiconductor, the bulk Fermi surface of the semiconductor is not relevant anymore. Indeed, both Enkovaara and our own computations show an absence of band gap in the density of states of the sandwiched silicon layer. Therefore, the system behaves like a metal/metal rather than a metal/semiconductor system. In his paper, Enkovaara [11] remarks that the overall electronic structure of the Si spacer layer can be described by the quantum well model and suggests that as a consequence the two layers coupling oscillations should also be observed with a diamond silicon spacer. Our experimental results as well as our computations confirm this suggestion.

Even if the overall behavior of the magnetic properties of the samples is consistent with the *ab initio* computations, the detailed magnetic behavior shows features that are difficult to understand within the available theoretical approaches. First of all, the coupling strength does not, in the range under investigation, decrease significantly with the spacer thickness. Indeed, in the case of a metallic spacer the coupling strength should decrease as the square of the spacer thickness while it should decrease exponentially in the case of an insulator or a semiconductor. However, the Si thickness experimentally explored is probably not large enough to properly determine the coupling decay length since, as already mentioned above, even small atomic scale Si thickness inhomogeneities will most probably average out the coupling strength and therefore artificially increase the decay length. In particular, this will strongly suppress any steep coupling strength increase for small Si thickness. Nevertheless, it can be noted that in semiconductors the electronic length scales are much larger than in metals and might therefore increase the coupling strength decay length compared to the one observed with metallic spacers. The second surprising feature is that the coupling strength seems not to vary with the temperature. This is a kind of hybrid behavior because for a metal it should decrease with temperature, whereas it should increase with temperature for a semiconductor spacer.

In this work we are showing that low temperature deposition (90 K) of Co/Si multilayers allowed the observation of saturation field oscillations consistent with the coupling oscillations predicted by *ab initio* computations. In addition, our experimental results show unusual

behaviors that are not understood yet. From a fundamental point of view, our results will renew the interest for ferromagnetic-metal/semiconductor systems because we are showing that it is possible to elaborate these artificial systems with low interfacial mixing. New effects are still to be evidenced in these systems. They may lead also, in the very near future, to new technological achievements in the spin electronics field.

*Corresponding author.
Email address: christian.meny@ipcms.u-strasbg.fr

[1] *Handbook of Nano-Technology*, edited by B. Bhushan (Springer-Verlag, Berlin, 2004).
[2] S. Ohnishi, A. J. Freeman, and M. Weinert, Phys. Rev. B **28**, 6741 (1983).
[3] P. F. Carcia, A. D. Meinhaldt, and A. Suna, Appl. Phys. Lett. **47**, 178 (1985).
[4] P. Grünberg, R. Schreiber, Y. Pang, M. B. Brodsky, and H. Sowers, Phys. Rev. Lett. **57**, 2442 (1986).
[5] 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).
[6] J. S. Moodera, L. R. Kinder, T. M. Wong, and R. Meservey, Phys. Rev. Lett. **74**, 3273 (1995).
[7] J. M. Fallon, C. A. Faunce, and P. J. Grundy, J. Appl. Phys. **88**, 2400 (2000).
[8] R. R. Gareev, D. E. Bürgler, M. Buchmeier, D. Olligs, R. Schreiber, and P. Grünberg, Phys. Rev. Lett. **87**, 157202 (2001).
[9] R. R. Gareev, D. E. Bürgler, M. Buchmeier, R. Schreiber, and P. Grünberg, J. Magn. Magn. Mater. **240**, 235 (2002).
[10] H. C. Herper, P. Weinberger, L. Szunyogh, and C. Sommers, Phys. Rev. B **66**, 064426 (2002).
[11] J. Enkovaara, A. Ayuela, and R. M. Nieminen, Phys. Rev. B **62**, 16 018 (2000).
[12] C. Quirós, J. I. Martín, L. Zárate, M. Vélez, and J. M. Alameda, Phys. Rev. B **71**, 024423 (2005).
[13] T. Luciński, P. Wandziuk, F. Stobiecki, B. Andrzejewski, M. Kopcewicz, A. Hütten, G. Reiss, and W. Szuskiewicz, J. Magn. Magn. Mater. **282**, 248 (2004).
[14] N. Yaacoub *et al.* (to be published).
[15] P. Panissod, J. P. Jay, C. Meny, M. Wojcik, and E. Jedryka, Hyperfine Interact. **97–98**, 75 (1996).
[16] P. Bruno, Phys. Rev. B **52**, 411 (1995).
[17] J. Faure-Vincent, C. Tiusan, C. Bellouard, E. Popova, M. Hehn, F. Montaigne, and A. Schuhl, Phys. Rev. Lett. **89**, 107206 (2002).
[18] V. Drchal, J. Kudrnovsk, and I. Turek, Comput. Phys. Commun. **97**, 111 (1996).
[19] I. Turek, V. Drchal, J. Kudrnovsk, and P. Weinberger, *Electronic Structure of Disordered Alloys, Surfaces and Interfaces* (Kluwer, Boston-London-Dordrecht, 1997).
[20] O. K. Andersen, Phys. Rev. B **12**, 3060 (1975).
[21] S. H. Vosko, L. Wilk, and M. Nusair, Can. J. Phys. **58**, 1200 (1980).