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Mimicking of a strong biquadratic interlayer exchange coupling in Fe/Si multilayers

J. Kohlhepp
Philips Research Laboratories, Prof. Holstlaan 4, 5656 AA Eindhoven, The Netherlands
and Department of Physics, Eindhoven University of Technology (EUT), 5600 MB Eindhoven, The Netherlands

M. Valkier and A. van der Graaf
Interfaculty Reactor Institute, TU Delft, 2629 JB Delft, The Netherlands

F. J. A. den Broeder
Philips Research Laboratories, Prof. Holstlaan 4, 5656 AA Eindhoven, The Netherlands

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The antiferromagnetic interlayer exchange coupling properties of sputtered Fe/Si multilayers have been studied by magnetometry and spin-polarized neutron reflectometry. Both the degree of antiferromagnetic alignment of adjacent ferromagnetic layers at zero field and the strength of the coupling are found to depend on the position in the multilayer stack. It is shown that these interlayer coupling variations are able to imitate an apparent strong biquadratic coupling. [S0163-1829(97)50102-0]

The discoveries of an antiferromagnetic (AF) exchange coupling between two ferromagnetic (FM) films across a nonmagnetic metallic interlayer,[1] the giant magnetoresistance (GMR) effect associated with such a coupling,[2] and the oscillatory behavior of the magnetic coupling[3] have initiated a huge amount of both experimental and theoretical investigations during the past decade. Whereas in metal/metal multilayers (ML's) the coupling is understood quite well,[4] the situation for ML's with semiconducting spacers is still unclear. Soon after the discovery of a very weak oscillatory exchange coupling between Fe layers across amorphous Si deposited at low temperature,[5] a very strong nonoscillatory AF coupling has been observed in sputtered Fe/Si MLs.[6] In this case the interlayer was found to be a crystalline interdiffused FeSi alloy. Subsequently it was concluded that the AF coupling could be converted to FM coupling by cooling to low temperature, at which the AF coupling could surprisingly be restored by light irradiation.[7] It was suggested that charge carriers, which are excited thermally or optically from the valence band, or alternatively from localized impurity states in the narrow band gap of a semiconducting ε-FeSi interlayer, mediate the exchange coupling.[7,8] Recently, however, we have shown that the AF exchange coupling strength in sputtered Fe/Si(FeSi) ML's strongly increases with decreasing temperature.[9] This is in clear contrast to the expected behavior of the excitation mechanism, where a decrease of the temperature should result in a weakening of the AF coupling strength.

Realizing the *negative* temperature coefficient of the AF coupling strength, the original supporters of the excitation mechanism have very recently reinterpreted their earlier temperature-dependent results[10] and performed additional experiments.[11] Based on a temperature-dependent analysis of the measured hysteresis loops[10] or the saturation fields H_S,[11] both groups concluded that a strongly temperature-dependent biquadratic-type coupling component (favoring a 90° alignment of adjacent FM layers) is responsible for the appearance of a remanent magnetization at low temperatures. Originally, such a biquadratic-type coupling has been discovered by domain microscopy studies of Fe/Cr/Fe trilayers where the 90° alignment of the Fe films at certain Cr thicknesses was *directly* observed.[12] Quite apart from the fact that the origin of such a high biquadratic contribution concluded to be present in Fe/Si(FeSi) ML's is still unclear, an *indirect* and *quantitative* determination of biquadratic coupling by simply fitting the magnetization loops of magnetic multilayers seems to be at least questionable.

The present paper is concerned with the nature of the apparent strong biquadratic interlayer exchange observed in sputtered Fe/Si ML's. By means of the magneto-optical Kerr effect (MOKE) and spin-polarized neutron reflectometry (PNR) measurements we were able to establish a strong dependence of the coupling characteristics on the position in the multilayer stack. It is mainly this striking phenomenon which causes the occurrence of remanences and pronounced rounding offs in the measured magnetizations loops and which is thus able to *mimic* a strong biquadratic interlayer exchange.

The Fe/Si ML's were prepared by dc-magnetron sputtering ($p_{\mathrm{base}} < 5\times10^{-5}$ Pa) at ~50 °C onto thermally oxidized silicon (oxi-Si) at an Ar pressure of 1 Pa. To allow optical access from the substrate side to the magnetic ML's, in some cases glass substrates have been used. For the experiments described here the individual Fe layer thicknesses were fixed at $t_{\mathrm{Fe}}=30$ Å whereas the nominal Si thicknesses varied between 8 and 40 Å in the $(t_{\mathrm{Fe}}\ \mathrm{Fe}/t_{\mathrm{Si}}\ \mathrm{Si})_N$ multilayers, where N is the number of bilayers and $t_{\mathrm{Fe}}(t_{\mathrm{Si}})$ are the nominal Fe(Si) thicknesses. No structural differences were found at all between the growth on glass and on oxi-Si, respectively. In low-angle x-ray diffracton (XRD) measurements (Cu K_α) Bragg peaks were observed up to $2\vartheta\simeq10°$ for all Si thicknesses, which is a clear indication of well-defined layering with smooth interfaces. However, the measured multilayer period Λ was considerably shorter than the actually expected sum of the individual Fe and Si thicknesses deduced from low-angle XRD measurements on calibration

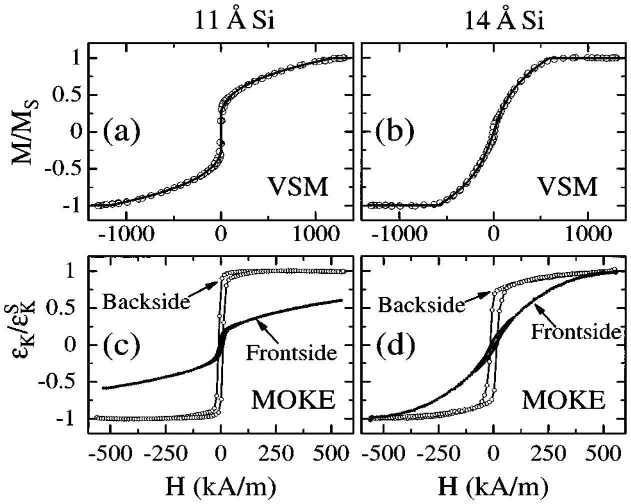

FIG. 1. Normalized hysteresis loops for a (30 Å Fe/11 Å Si$)_{20}$ (left part) and a (30 Å Fe/14 Å Si$)_{20}$ multilayer (right part) grown on glass, measured with VSM and longitudinal MOKE at 295 K. The thick solid lines in the VSM loops [(a) and (b)] are fits resulting in $J_1 = -0.679$ and $J_2 = -0.398$ mJ/m^2 for 11 Å Si and $J_1 = -0.504$ and $J_2 = -0.098$ mJ/m^2 for 14 Å Si.

samples. Such a contraction of Λ results from a large decrease of the Si atomic volume in an Fe environment caused by iron-silicide formation. In agreement with earlier growth studies on sputtered Fe/Si multilayers, our own high-angle XRD measurements confirmed a complete $(110)_{\text{bcc}}$-textured growth with typical perpendicular coherence lengths around 200 Å for a nominal t_{Si} up to 16 Å. Above $t_{\text{Si}} = 16$ Å an amorphization of the interlayer starts, indicated by a distinct broadening of the (110) main peak corresponding to a coherence length limited by a single Fe layer and the disappearence of high-angle satellite reflections. The most probable explanation for the crystalline interlayer growth below $t_{\text{Si}} = 16$ Å is the aforementioned iron-silicide formation during growth. Especially the nonmagnetic CsCl-structure (B2) FeSi phase seems to be a good canditate for the crystalline interlayer due to its excellent lattice match to bulk bcc-iron. Recently, it has been shown that this metastable silicide phase can be stabilized in thin films.[13] B2-FeSi is metallic[14] and is now held responsible for mediating the interlayer exchange coupling between adjacent Fe layers in Fe/Si-based systems.[15]

Besides the appearance of a remanent magnetization another striking feature is observed in the hysteresis loops of antiferromagnetically coupled Fe/Si ML's.[9,10,16] They exhibit a distinct convex shape [see Figs. 1(a) and 1 (b)]. Phenomenologically this behavior can be described by the presence of a so-called biquadratic coupling term E_{bq}, favoring a 90° alignment of adjacent FM layers, in addition to the bilinear Heisenberg-type AF coupling term E_{bl} (180° alignment) in the interlayer exchange energy:[12]

$$E_{\text{ex}} = E_{\text{bl}} + E_{\text{bq}} = -J_1 \cos(\Delta\Theta) - J_2 \cos^2(\Delta\Theta). \qquad (1)$$

Here $\Delta\Theta$ is the angle between two adjacent FM layers and J_1 (J_2) is the bilinear (biquadratic) coupling parameter. Theoretical magnetization loops can be calculated by simply minimizing the total magnetic energy of the multilayers. A least-squares fit of the calculated to the measured loops thus gives J_1 and J_2. For the majority of our experiments we could indeed find suitable combinations of J_1 and J_2 which were able to reproduce the remanences and the loop shapes in a satisfactory manner [see for example Figs. 1(a) and 1(b)].

Various possible mechanisms causing a biquadratic contribution have been proposed recently. But neither intrinsic interlayer properties,[17] nor extrinsic factors such as interlayer thickness fluctuations in connection with short-period AF-FM oscillations,[18] (super)paramagnetic impurities (''loose spins'') in the interlayer or at the interface to the FM layers,[19] nor magnetic-dipole fields created by magnetic layers with uncorrelated roughness[20] are able to explain the strength and the strong temperature dependence of J_2 observed in our experiments.

We can resolve this puzzle by introducing an alternative mechanism which is responsible to contribute at least partly to the rounding of the loop shapes and the occurrence of remanence, namely a dependence of the exchange coupling properties on the position in the ML stack. A first hint for the existence of such a phenomenon is given by studies of ML's with different numbers of bilayer repetitions N.[16,21] These showed a remarkable decrease of the remanent magnetization from nearly 90% for an Fe/Si/Fe sandwich to almost zero for $N > 60$. Additionally, a change from a strongly convex curved magnetization loop to a more linear one, characteristic for almost pure bilinear coupling, was observed. In order to check the variation of the coupling characteristics with the position in the multilayer stack in more detail, we took advantage of the limited information depth of MOKE to measure loops both from the surface (front side) and from the glass substrate side (back side), independently. Figure 1 shows the results of such an investigation for two different ML's with 11 and 14 Å nominal Si thicknesses. Here, the vibrating-sample magnetometer (VSM) loops could be fitted excellently with the bilinear/biquadratic coupling model, although the longitudinal MOKE loops definitely show that both the degree of ferromagnetic alignment of adjacent FM layers and the rotation process depend on the position in the ML stack. At the bottom of the ML, close to the substrate, the FM layers are weakly AF coupled, while the high remanence is indicative for a very substantial degree of FM coupling. On the other hand, the top layers show a strong AF coupling with almost no remanence.

In order to support our MOKE observations we additionally performed polarized neutron reflectivity (PNR) measurements at the time-of-flight reflectometer of the Interfaculty Reactor Institute (IRI) in Delft.[22] In our PNR experiments the reflected neutron intensity was measured as a function of the incident neutron wave-vector component normal to the sample surface, $q_0 = 2\pi \sin\vartheta/\lambda$, where λ is the neutron wavelength, and ϑ is the angle of incidence, and as a function of the polarization state of the incident neutron beam with respect to an applied magnetic field. Unfortunately, with the experimental setup an exit-beam polarization analysis was not possible.

Shown in Fig. 2 are reflectivity data taken at room temperature in three different fields applied parallel to the surface of a (30 Å Fe/14 Å Si$)_{20}$ ML with the incident neutron beam polarized parallel (+) or antiparallel (−) to the field.

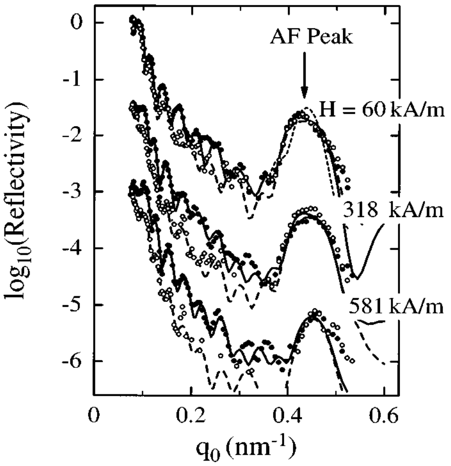

FIG. 2. Polarized neutron reflectivities for a $(30\ \text{Å Fe}/14\ \text{Å Si})_{20}$ multilayer in different applied magnetic fields H at room temperature. The solid (open) symbols are the measured and the solid (long-dashed) lines are the calculated reflectivities for the neutron spin parallel (antiparallel) to the field. Only the incident beam was polarized and spin-flip scattering was neglected in the calculations. The short dashed curve (60 kA/m) is the calculated reflectivity (neutron spin parallel) at the AF peak for a magnetic order limited only by the structural coherence length.

The most important feature in the low-field spectrum is the presence of a strong Bragg reflection at $q_0 \simeq 0.44\ \text{nm}^{-1}$, which corresponds to a magnetic periodicity Λ_{mag} twice the chemical modulation length Λ deduced from low- and high-angle XRD. Such a Bragg peak is expected for an AF ordering of subsequent FM layers and thus confirms the existence of an AF exchange coupling across the FeSi interlayer in our sample. The drop of the intensity of the AF Bragg peak observed with increasing field strength is simply the effect of an enhanced Zeeman energy in competition with the coupling energy, causing a decrease of AF alignment and consequently a decrease of the magnetic contrast between adjacent FM layers. Remarkable is the considerable width of the AF Bragg peak, which is a *clear sign* of a poor magnetic coherence length and thus an additional indication of a strongly depth-dependent coupling behavior along the ML. For a magnetic order limited by the structural coherence of the ML one would expect much narrower AF peaks, which is exemplarily shown for one field (60 kA/m) and one spin direction (+) by the short-dashed curve in Fig. 2.

However, due to the missing exit-beam polarization analysis in our setup, a separation between spin-flip and non-spin-flip scattering was not possible. Thus, vector magnetometry could not be performed and quantitative statements about the magnetic structure of the ML cannot be made. Nevertheless, with the help of some simplifications concerning the magnetic structure, at least some *qualitative* conclusions can be drawn. In the following it is assumed that the magnetic moments of the magnetic layers are always aligned parallel or antiparallel to the applied field. Therefore only perfect FM or AF aligned regions (domains) are allowed between the adjacent FM layers, and consequently spin-flip scattering cannot take place. The reflectivities depend on the scattering length density (SLD), $\Gamma(z)$, which in absence of spin-flip scattering can be written as $\Gamma_{\pm}(z) = \Gamma_n(z) \pm \Gamma_m(z) = n_0(z)(b(z) \pm p(z))$, with n_0 the atomic number density, b the nuclear and p the magnetic scattering length. Here $\Gamma_n(z)$ is the nuclear SLD and $\pm\Gamma_m(z)$ is the magnetic SLD which is proportional to the in-plane average magnetization parallel $(+)$ or antiparallel $(-)$ to the neutron spin.

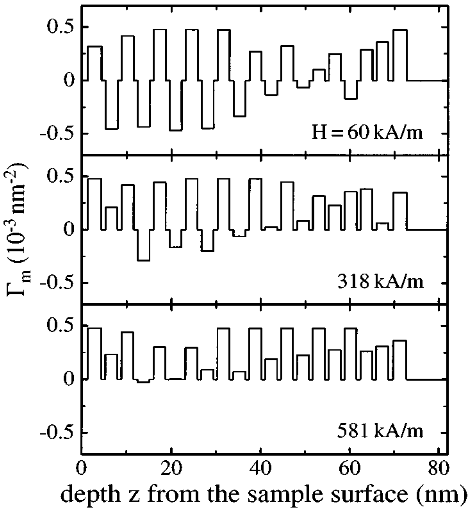

FIG. 3. Magnetic scattering length density (Γ_m) profiles corresponding to nonspin-flip reflectivity calculations of Fig. 2. In the framework of our model $\pm\Gamma_m$ is proportional to the magnetization of a layer which lies parallel $(+)$ or antiparallel $(-)$ with respect to the applied field.

The theoretical reflectivity was calculated using the so-called matrix method,[23] and convoluted with the experimental resolution. For this purpose, the sample was divided into slabs alternatingly composed of 9 Å of a nonmagnetic iron silicide with a Γ_n very close to $Fe_{50}Si_{50}$ and 27 Å of an iron-rich $Fe_{100-x}Si_x$ $(x<15)$ magnetic alloy (for more details see Ref. 24). Keeping this nuclear SLD profile of the ML constant, the measured reflectivities for the different magnetic field values were fitted by adjusting the magnetization profile of the sample. Figure 3 shows the resulting magnetic non-spin-flip SLD profiles exemplarily for the PNR spectra of Fig. 2. Here, the solid (dashed) lines in Fig. 2 represent the optimum fits of the calculated reflectivities to the measured data points (symbols) for the neutron spin parallel (antiparallel) to the applied magnetic field. Inspecting in Fig. 3 the low-field (60 kA/m) $\Gamma_m(z)$ profile, a strong dependence of the magnetic alignment of FM layers on the position in the ML becomes evident. While the magnetizations of the top layers show a nearly perfect AF alignment, the coupling in the bottom half of the sample is predominantly FM, which is in agreement with the interpretation of our MOKE measurements. Applying an increasing external field (see for instance $H = 318$ and 581 kA/m in Fig. 3) results in an increase of the magnetizations parallel to the field direction. But once again, this process is not homogeneous, it is in turn depending on the position in the ML stack. It is obvious that one needs much more field to achieve a magnetic saturaton for layers close to the sample surface than for those in the bottom half of the ML. Thus, not only the zero-field alignment, but also the coupling strength is depth dependent.

All in all, we have measured the PNR reflectivities and

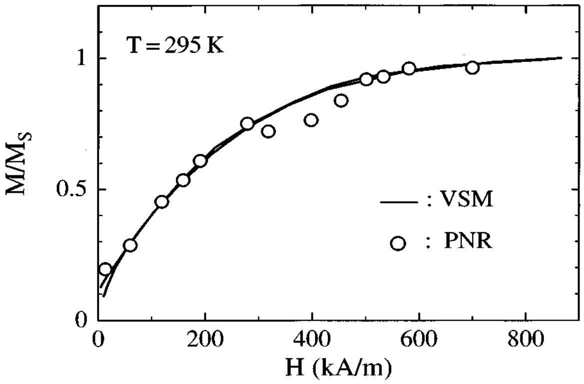

FIG. 4. Comparison of the normalized magnetization loops of a (30 Å Fe/14 Å Si)$_{20}$ multilayer measured by vibrating sample magnetometry (VSM) and polarized neutron reflectivity (PNR) experiments.

subsequently determined the $\Gamma_m(z)$ profiles for 13 different magnetic fields. Correspondingly, an averaging of the magnetizations in the magnetic SLD profiles as a function of the applied magnetic field provides us with the magnetization loop of the sample. Figure 4 shows the loops deduced from our PNR study and measured by (depth-insensitive) vibrating sample magnetometry. The excellent matching of the PNR data with the VSM curve is striking and supports at least in a qualitative way the validity of our depth-dependent model, despite the serious simplifications which have been made.

A depth-dependent coupling provides a natural explanation for the experimental observations without being forced to introduce an unusual high biquadratic coupling term in the energetics. Since each FM layer can be described by a magnetization loop with a remanence and a saturation field characteristic for the position in the ML, a superposition of all these individual loops results in macroscopic hysteresis curves with FM fractions and nonlinear slopes. A variation of the interlayer exchange coupling in the vertical direction can thus imitate an apparent strong biquadratic interlayer exchange and can lead to serious misinterpretations of experimental results.

Naturally, the question arises as to which mechanism causes this inhomogeneous coupling behavior. Probably, the growth of the first few layers on the amorphous substrate is strongly disturbed, resulting in local ferromagnetic short circuits, which strongly influence the loop shapes and the coupling strength.[25] This interpretation is supported by an additional experiment where the first Fe/Si layers have been replaced by a Cr/Si ML with an identical growth behavior.[16] A single Fe/Si/Fe sandwich grown on top of such a simulated Fe/Si base ML already showed a predominantly AF coupled magnetization loop. Besides usual pinholes, as for instance grain boundaries filled with a ferromagnetic phase, also chains of nearest-neigbor Fe atoms in the not perfectly ordered FeSi interlayer which percolate the magnetization between adjacent Fe layers are able to mediate FM coupling. In the present case especially, these percolation bridges may become successively ferromagnetic on cooling, explaining the observed remarkable temperature dependences of the remanences and the loop shapes.

In conclusion, we have shown that the degree of antiferromagnetic alignment of adjacent ferromagnetic layers and the coupling strength definitely depend on the position in the Fe/Si ML stack. Consequently, vertical as well as lateral variations in the bilinear AF interlayer coupling and FM coupling are able to imitate an apparent strong temperature-dependent biquadratic coupling and thus seriously obstruct the interpretations of *real* biquadratic contributions to the exchange coupling in magnetic ML's.

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