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Biquadratic interlayer exchange coupling in epitaxial Fe/Si/Fe

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We have studied the biquadratic exchange coupling in epitaxially grown Fe/Si/Fe. The temperature and thickness dependence of the biquadratic coupling strength were determined unambiguously by fitting the easy- and hard-axis magneto-optical Kerr effect loops. The origin of the biquadratic coupling can be fully understood in terms of Slonczewski's loose spins mechanism. © *2000 American Institute of Physics.* [S0021-8979(00)31208-7]

I. INTRODUCTION

The bilinear interlayer exchange coupling in Fe/Si/Fe layers is rather exceptional, in that its strength decays exponentially with the Si spacer layer thickness, in contrast to the "normal" oscillatory behavior.[1,2] The origin of this bilinear coupling can be explained by the formation of a metallic iron-silicide spacer with the CsCl structure that has a high density of states peak above the Fermi level, and can be described in terms of the Bruno electron-optics model[3] with imaginary extremal Fermi vectors, or by the Anderson *sd*-mixing model.[4]

On the other hand, the mechanism behind the biquadratic exchange coupling observed in Fe/Si-based layers has not been clearly understood up to now. Several attempts have been made to clarify its origin by analyzing the temperature dependence of the biquadratic coupling strength in Fe/Si multilayers.[5–9] However, in these multilayers the "true" biquadratic coupling is obscured by vertical and lateral variations of the coupling strengths.[10] Therefore, no definite conclusions about the origin of the biquadratic coupling can be drawn from the previous studies on Fe/Si multilayers.

In this paper, we present a study of the biquadratic exchange coupling in well-defined epitaxially grown Fe/Si/Fe trilayers. The magnetization was analyzed with the magneto-optical Kerr effect (MOKE) to avoid lateral variations as much as possible. Since these layers contain only one spacer, there are no vertical variations. We will show that the origin of the "true" biquadratic coupling can be understood in terms of Slonczewski's loose spins mechanism.

II. EXPERIMENTAL RESULTS

The Fe/Si/Fe layers were grown at room temperature in a multichamber molecular beam epitaxy (MBE) system (VG-Semicon V80M), with a base pressure better than 2×10^{-11} mbar. An electron-gun source with feedback control of the flux was used for the deposition of Fe whereas Si was evaporated from a temperature stabilized Knudsen cell. All thicknesses were controlled by calibrated quartz-crystal monitors. The layers were grown on Ge(001) substrates, which were cleaned by an Ar^+ sputter and anneal treatment prior to deposition. In a previous study, we have shown that in these Fe/Si/Fe layers the Si spacer transforms to metallic c-$Fe_{1-x}Si$ by Fe and Si interdiffusion,[11] which leads to an approximately 6% increase of the nominal spacer thickness. This correction is rather small, and therefore we will refer to the nominal layer thicknesses in the rest of this paper. Biquadratic coupling was studied in a number of samples. Uniform samples with the following composition: Ge(001)+60 Å Fe $+t_{Si}$Si+45 Å Fe+30 Å Si, with t_{Si}= 14, 14.5, 15, 15.25, 16, and 16.25 Å, and wedge-shaped samples composed of: (wedge A) Ge(001)+115 Å Fe+8-18 Å Si-wedge+90 Å Fe+30 Å Si, and (wedge B) Ge(001)+60 Å Fe+7-17 Å Si-wedge+45 Å Fe+30 Å Si.

The interlayer coupling constants were determined by analyzing the magnetization hysteresis curves of the Fe/Si/Fe layers. As an example, the room temperature [100] easy-axis and [110] hard-axis MOKE loops of wedge A, for nominal Si thicknesses of 12.4 and 13.7 Å, are shown in Fig. 1. The magnetization hysteresis curves can be quantitatively described by considering the expression for the total areal energy density of the two magnetic layers, which reads

$$
\begin{aligned}
E = & -\mu_0 M_s H[t_1 \cos(\phi_1 - \phi_H) + t_2 \cos(\phi_2 - \phi_H)] \\
& + K t_1 \cos^2(\phi_1)\sin^2(\phi_1) + K t_2 \cos^2(\phi_2)\sin^2(\phi_2) \\
& - J_1 \cos(\phi_1 - \phi_2) - J_2 \cos^2(\phi_1 - \phi_2),
\end{aligned} \tag{1}
$$

with M_s the saturation moment of layer 1 and 2 with thickness t_1 and t_2. $M_s t_1$ and $M_s t_2$ were determined by SQUID (superconducting quantum interference device) magnetization measurements. Here ϕ_1 and ϕ_2 are the angles between the magnetization of layers 1 and 2 and the [100] easy axis, respectively, while ϕ_H is the angle between the field H and the [100] axis ($\phi_H = 0$ for easy-axis loops and $\phi_H = 45°$ for hard-axis loops). The cubic anisotropy constant K is taken equal for layers 1 and 2.

J_1 is the bilinear coupling constant (<0 for antiferromagnetic coupling) and J_2 is the biquadratic coupling constant (<0 for 90° coupling). Due to the finite penetration depth of the incident laser beam only part of the bottom Fe layer contributes to the MOKE signal, which can lead to a negative remanence, although the bottom Fe layer is thicker than the top one. By combining SQUID and MOKE magne-
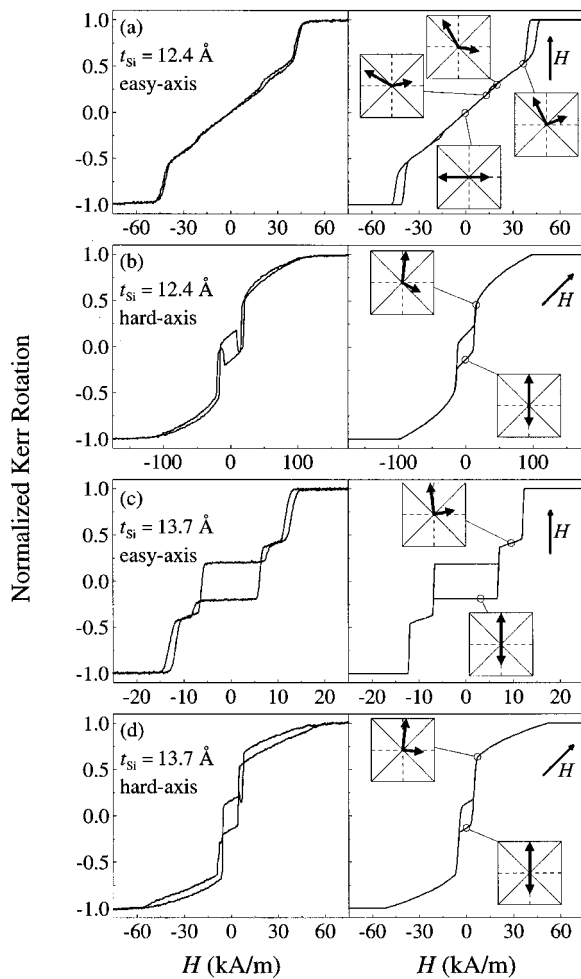
FIG. 1. Left-hand side: Measured MOKE loops at room temperature of (a), (b) Ge(001)+115 Å Fe+12.4 Å Si+90 Å Fe+30 Å Si, and (c), (d) Ge(001) +115 Å Fe+13.7 Å Si+90 Å Fe+30 Å Si with the field applied along the easy and hard axes as indicated. Right-hand side: Fits of the loops with Eq. (1). The insets show the orientations of the magnetization vectors at several field values; dashed lines are easy axes and solid lines are hard axes.

FIG. 2. Temperature dependence of J_2 for Ge(001)+60 Å Fe $+t_{Si}$Si+45 Å Fe+30 Å Si, with t_{Si} indicated. The solid lines are fits to the experimental data with the loose spins model, as explained in the text.

tization loops we have determined that only 50% of the bottom Fe layer contributes to the observed Kerr rotation.

For a Si thickness of 12.4 Å, J_1 and J_2 are of the same order of magnitude as Kt_1 and Kt_2. We therefore adopted an energy minimum path approach to Eq. (1) to fit the hysteresis curves of Figs. 1(a) and 1(b), which correctly takes into account the competition between the coupling energy and the anisotropy when the magnetic moments rotate across the anisotropy barriers. The fits are shown on the right-hand side of Fig. 1 and the insets in the figure illustrate the various states of the magnetic moments as function of the applied field. Upon decreasing the field from saturation along the easy axis, the moments first jump to a nearly 90° orientation, after which they gradually rotate toward an antiparallel alignment, perpendicular to the field with zero remanence. When the field is decreased from saturation along the hard axis, first the moments gradually rotate to a nearly 90° orientation, after which a jump occurs toward a completely antiparallel alignment. We stress that both easy- and hard-axis loops are fitted consistently with one set of parameters, yielding J_1
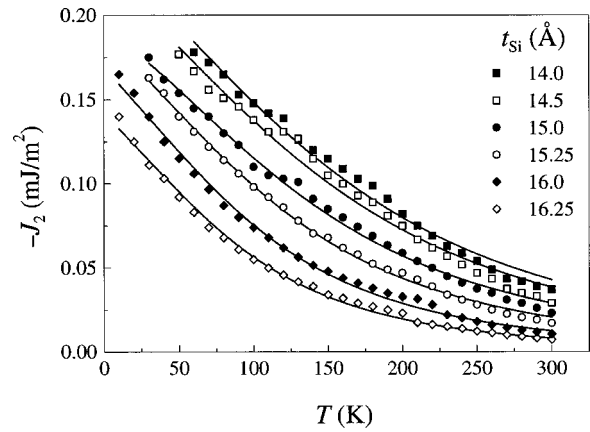
$= -0.515\,\text{mJ/m}^2$,　　$J_2 = -0.080\,\text{mJ/m}^2$,　　and　　$K = 3.2 \times 10^4\,\text{J/m}^3$.

For a Si thickness of 13.7 Å, shown in Figs. 1(c) and 1(d), the minimum path approach results in a large overestimation of the hysteresis, because J_1 and J_2 are smaller than Kt_1 and Kt_2 and only rotation of the magnetic moments is allowed, without thermal activation across the anisotropy barriers. It is therefore more appropriate to use an absolute minimum energy approach to Eq. (1) in this case. For the easy-axis loops two jumps can be observed: one from saturation toward a 90° orientation of the moments, and a second jump to the antiparallel alignment. The hard-axis loop shows the same features as Fig. 1(b). Again, both easy- and hard-axis loops are fitted with one set of parameters, yielding J_1 $= -0.160\,\text{mJ/m}^2$,　　$J_2 = -0.028\,\text{mJ/m}^2$,　　and　　$K = 3.2 \times 10^4\,\text{J/m}^3$ for this Si thickness.

In this way the magnitude of J_1 and J_2 of the six uniform samples was determined from room temperature down to 10 K. We note that for these films with nominal Fe layer thicknesses of 60 and 45 Å the anisotropy K ranges between $1.8 \times 10^4\,\text{J/m}^3$ at room temperature and $3.5 \times 10^4\,\text{J/m}^3$ at 10 K. J_1 is always <0 (antiferromagnetic coupling; see Fig. 3) and increases only slightly with decreasing temperature, in agreement with earlier observations.[8] In the present paper we want to focus on the biquadratic coupling, whose temperature dependence is plotted in Fig. 2. As can be seen J_2 increases remarkably strongly with decreasing temperature for all Si thicknesses.

III. DISCUSSION

There are a number of possible mechanisms that can account for the observed biquadratic exchange in these Fe/ Si-based layers. First of all, J_2 may be an intrinsic higher-order term of the coupling, as was claimed recently.[9] We rule out this possibility, because the magnitude of an intrinsic second-order coupling term J_2 is generally orders of magnitude smaller than J_1 and its temperature dependence less dramatic.

Theoretically, a 90° coupling can also arise from thickness fluctuations of the spacer layer, which cause a compe-
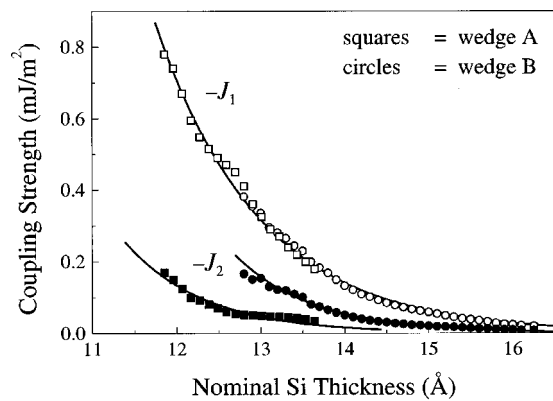
FIG. 3. Thickness dependence of J_1 (open symbols) and J_2 (closed symbols) at room temperature.

tition between ferromagnetic and antiferromagnetic coupling for neighboring regions.[12] For Fe/Si, however, the bilinear coupling J_1 always favors an antiparallel alignment of the magnetic layers and therefore lateral thickness variations do not lead to a frustration of the coupling here, ruling out this possibility as well.

The remaining mechanism is biquadratic coupling mediated by paramagnetic atoms or (super)paramagnetic clusters of atoms in the spacer layer, as was proposed by Slonczewski.[13] These so-called ''loose spins'' can couple to both ferromagnetic layers via the indirect exchange, which is also responsible for J_1. $J_2(T)$ can be expressed in terms of an areal loose spins density c and the total interaction potential U between loose spins and ferromagnetic layers. For a complete description of the loose spins model, see Ref. 13.

Unlike the other mechanisms for biquadratic coupling, the loose spins model does predict a strong temperature dependence of J_2. Figure 2 is supplemented with fits of $J_2(T)$ with the loose spins model. The areal loose spins density c and the interaction potential U were adjusted for the fit. The density of loose spins converged consistently to approximately 1% for all thicknesses, and $U/k_B = 343$, 334, 292, 266, 222, and 199 K for $t_{Si} = 14$, 14.5, 15, 15.25, 16, and 16.25 Å, respectively.

A separate check for the biquadratic coupling resulting from loose spins follows from the spacer layer thickness dependence of J_1 and J_2, as shown in Fig. 3 for wedges A and B. Because the interaction potential U is directly related to J_1, both J_1 and J_2 should have the same thickness dependence. Indeed, as can be seen in Fig. 3, both J_1 and J_2 decay

exponentially as function of the spacer layer thickness with approximately the same decay length.

Note the consistency between wedges A and B in the magnitude of J_1, demonstrating that J_1 does not depend critically on the exact spacer layer structure, that may have some deposition run-to-run variations. This is in agreement with the explanations for the antiferromagnetic coupling in Fe/Si/Fe, resulting from a high density of states peak above the Fermi level that is present for stoichometric and also for defective c-FeSi.

On the other hand, J_2 of wedge A differs considerably in magnitude from wedge B. This corroborates our interpretation with the loose spins model, because J_2 is very sensitive to the exact loose spins concentration, apparently different for our two wedges, which were grown in separate deposition runs, also with different Fe layer thicknesses. From the temperature dependence of the bilinear coupling strength, it seems that the J_1 resulting from the loose spins is negligible compared to the J_2. Although the reason for this is not completely clear up to now, we believe that the loose spins J_1 is averaged out due to a distribution in the strength of the interaction U between the loose spins and the magnetic layers.

IV. CONCLUSIONS

In conclusion, we have studied the biquadratic exchange coupling in well defined MBE-grown Fe/Si/Fe layers. In contrast to earlier studies we have determined the coupling parameters, not disturbed by vertical and lateral fluctuations of the coupling properties, as usually is the case for Fe/Si multilayers. Biquadratic coupling in Fe/Si/Fe can be understood in terms of Slonczewski's loose spins model.

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