# Origin of Biquadratic Exchange in Fe/Si/Fe

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The thickness and temperature dependences of the interlayer exchange coupling in well-defined molecular beam epitaxy-grown Fe/Si/Fe sandwich structures have been studied. The biquadratic coupling shows a strong temperature dependence in contrast to the bilinear coupling. Both depend exponentially on thickness. These observations can be well understood in the framework of Slonczewski's loose spins model [J. Appl. Phys. **73**, 5957 (1993)]. No bilinear contribution of the loose spins to the coupling was observed.

Recently, an exceptional type of coupling was found in Fe/Si/Fe, strongly antiferromagnetic (AF) and varying exponentially with the spacer layer thickness [1]. This rather unique behavior of the coupling is mediated by $c$-Fe$_{1-x}$Si formed by Fe diffusion into the Si spacer layer [2] and can be qualitatively understood in terms of the Bruno electronoptics model [3] with imaginary extremal Fermi vectors or by the Anderson $sd$-mixing model [4].

Apart from bilinear exchange coupling also a biquadratic contribution to the coupling was observed in sputtered Fe/Si multilayers [5–10]. However, a qualitative and quantitative interpretation of the biquadratic coupling in these *multilayers* is impossible due to lateral and vertical variations of the coupling properties [8,10]. Nevertheless, the Fe/Si system is unique and particularly attractive for studies on biquadratic coupling. Unlike the oscillatory RKKY interactions, the monotonically (exponentially) decaying intrinsic AF exchange coupling limits the possible mechanisms for biquadratic exchange and allows a flexible and large range of spacer thicknesses to be studied.

In this Letter we present a study of biquadratic exchange coupling in well-defined molecular beam epitaxy (MBE)-grown Fe/Si/Fe *trilayers* which do not suffer from the complications reported for multilayers. Based on combined detailed measurements of the temperature and thickness dependence of the bilinear as well as the biquadratic coupling constants, it is shown that biquadratic exchange in Fe/Si/Fe is due to a small concentration of (super)paramagnetic Fe clusters in the spacer layer, which act as so-called "loose spins." By exploiting the unique exponential character of the coupling, we are able to demonstrate that $J_1$ and $J_2$ are caused by the same interaction potential, which is a basic ingredient for modeling the loose spins coupling. Surprisingly, however, a contribution of the loose spins to the bilinear coupling seems to be absent.

The Fe/Si/Fe layers were grown at room temperature in a molecular beam epitaxy system (VG-Semicon V80M) with a base pressure of $2 \times 10^{-11}$ mbar. An $e$-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. The thicknesses were controlled by calibrated quartz crystal monitors. The layers were grown at room temperature on Ge(001) substrates, which were cleaned by several Ar$^+$ sputter (700 °C) and anneal (780 °C) treatments until a sharp Ge(001)-(2 × 1) LEED pattern was observed.

Previous studies have shown that in these Fe/Si/Fe trilayers the Si spacer transforms to metallic $c$-Fe$_{1-x}$Si by Fe and Si interdiffusion, and the whole stack grows epitaxially bcc(001)-like on the Ge(001) substrate [1,2]. However, this recrystallization only slightly alters the effective thickness of the spacer layer, and therefore we will refer to the nominal Si spacer layer thickness for the remainder of this Letter. The temperature dependence of the coupling was studied in six samples of 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. The spacer layer thickness dependence was measured in a wedge-shaped sample composed of Ge(001) + 60 Å Fe + 7–17 Å Si wedge + 45 Å Fe + 30 Å Si.

Figure 1 shows two normalized magneto-optical Kerr effect (MOKE) hysteresis curves with the field applied along the [100] easy axis, at 300 and 100 K for a nominal Si thickness $t_{Si}$ = 15.25 Å. These magnetization curves are representative for all of the other loops. At 300 K the magnetization loop can be characterized by two switching fields, indicated by $H_1$ and $H_2$ in the figure (defined in the middle of the hysteresis). Going from high to low fields, $H_1$ corresponds to a reorientation of the magnetic moments of the two Fe layers from a parallel to a perpendicular alignment, and at $H_2$ the alignment of the magnetizations changes from perpendicular to antiparallel. The difference between $H_1$ and $H_2$ is a measure of the biquadratic coupling strength, $J_2$, and the sum is characteristic for the bilinear coupling strength, $J_1$. When the biquadratic coupling strength is larger than the bilinear coupling strength, as is the case in Fig. 1(b) for 100 K, only one step is observed in the magnetization loop. The perpendicular alignment of the moments of the Fe layers is then maintained down to zero field. In the latter case $J_1$ and $J_2$ can be evaluated only from a fit of the experimental curves. We note
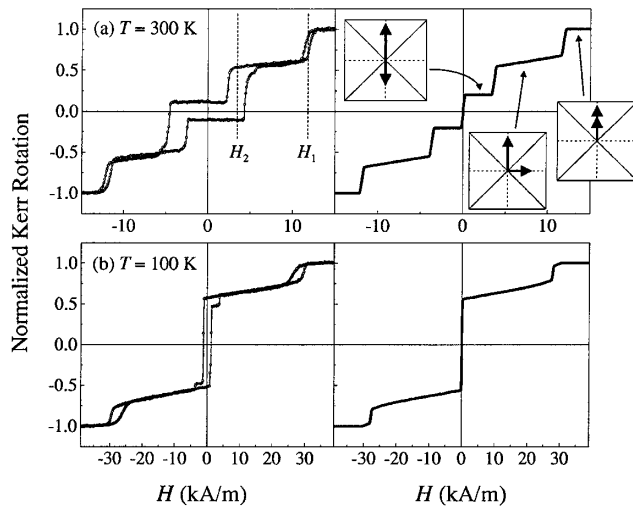
FIG. 1.   MOKE hysteresis curves for $t_{Si} = 15.25$ Å with the field along the [100] easy axis at (a) 300 K and (b) 100 K, and corresponding fits (right hand side) of the experimental curves with Eq. (1). The insets show the relative orientations of the magnetic moments for different fields (dashed lines are the easy axes and solid lines are the hard axes).

that sometimes small steps in the magnetization loops are observed at low fields, as also can be seen in Fig. 1(b), which are magneto-optical artifacts caused by a small misalignment of the easy axis with respect to the field.

The magnetization hysteresis curves can be fitted by considering the phenomenological expression for the areal energy density of the two magnetic layers

$$E = -\mu_0 M_s H[t_1 \cos(\phi_1 - \phi_H) + t_2 \cos(\phi_2 - \phi_H)]$$
$$+ Kt_1 \cos^2(\phi_1)\sin^2(\phi_1) + Kt_2 \cos^2(\phi_2)\sin^2(\phi_2)$$
$$- J_1 \cos(\phi_1 - \phi_2) - J_2 \cos^2(\phi_1 - \phi_2), \qquad (1)$$

with $M_s$ the saturation moment of layers 1 and 2 with thicknesses $t_1$ and $t_2$. Here $\phi_1$ and $\phi_2$ are the angles between the magnetization of layers 1 and 2 and the [100] easy axis, respectively, while $\phi_H$ is the angle between the field $H$ and the [100] axis. The cubic anisotropy constant $K$ was assumed equal for layers 1 and 2. An extra uniaxial anisotropy as is sometimes observed for epitaxial ultrathin Fe(001) films on semiconductors like Ge and GaAs (see, e.g., [11]), was not observed in the relatively thick Fe films [>30 ML (monolayer)] used in our experiments and thus neglected. $J_1$ is the bilinear coupling constant (<0 for AF coupling) and $J_2$ is the biquadratic coupling constant (<0 for 90° coupling). By a minimalization of Eq. (1) as function of the applied field $H$, the magnetization loops can be reproduced in a satisfactory way by choosing the correct combination $J_1$ and $J_2$ as is demonstrated on the right hand side of Fig. 1. Uncertainties in the determination of the coupling constants when $-J_2 > -J_1$ are overcome by combining easy and hard axes loops. The anisotropy $K$, evaluated from the shape of easy and hard axes loops, decreases with increasing temperature from about $3.5 \times 10^4$ J/m$^3$ at 10 K to ap-

proximately $1.8 \times 10^4$ J/m$^3$ at 300 K. The temperature dependences of $J_1$ and $J_2$ resulting from the fits are plotted in Figs. 2(a) and 2(b), respectively, for the six different nominal Si thicknesses.

$J_1$ is antiferromagnetic and decreases slowly with increasing temperature for all Si thicknesses. In a previous paper by de Vries *et al.* [1] it was shown that the origin of the exponential thickness dependence of $J_1$ can be understood within the Anderson $sd$-mixing model applied to Fe/Si by Shi *et al.* [12] or within the framework of Bruno's theory introducing the concept of an imaginary critical Fermi wave vector [3]. Both models predict a different temperature dependence. However, the temperature dependence of $J_1$ is rather small, and we cannot rule out completely that the small temperature dependence observed is a result of a decrease of the (surface) magnetization of the Fe layers. Nevertheless, at this point the temperature dependence of $J_1$ seems to be in favor of the $sd$-mixing model by Shi *et al.*, which predicts a decreasing coupling strength with increasing temperature, in contrast to Bruno's model.

The biquadratic coupling $J_2$, shown in Fig. 2(b), has a much stronger temperature dependence than $J_1$. There are a number of possible mechanisms for this biquadratic coupling. First of all, $J_2$ may be of intrinsic origin as was claimed recently [9]. We reject this possibility because the
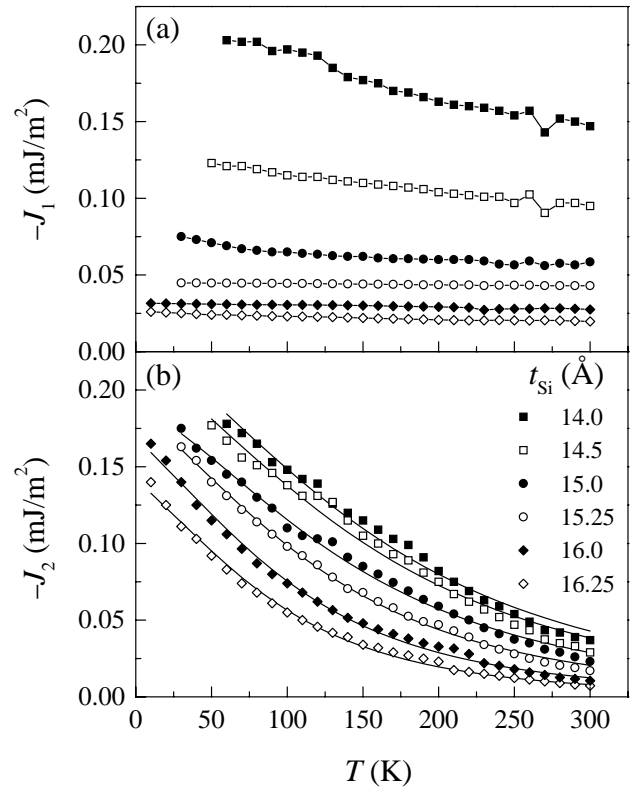


FIG. 2.   Temperature dependence of (a) $J_1$ and (b) $J_2$ for six Si thicknesses as indicated in the figure. The solid lines in (b) are fits to the experimental data with Eq. (5), as explained in the text.

magnitude of an intrinsic second order term $J_2$ is generally orders of magnitude smaller than $J_1$, much smaller than observed experimentally [3]. Furthermore, the temperature dependence should be less dramatic. Two other extrinsic mechanisms for biquadratic coupling are proposed by Slonczewski.

The fluctuation mechanism for biquadratic exchange [13] predicts a $J_2$ when spatial fluctuations of the interlayer thickness cause a competition between ferromagnetic and antiferromagnetic coupling for neighboring regions. The resulting frustration can lead to a perpendicular alignment of the magnetic moments, when the size of the thickness fluctuations is below the size of the domain walls. 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 [14]. Furthermore, the fluctuation mechanism predicts a temperature dependence $J_2(T) \propto [J_1(T)]^2$. As shown in Fig. 2(a), $J_1$ is decreased only by 20% to 30% at 300 K compared to 10 K, while $J_2$ decreases by a factor of 19, ruling out an interpretation in terms of the fluctuation model. We also note that magnetic dipole fields created by roughness can result in a biquadratic alignment of the moments [15]. The magnitude of this contribution is however small.

The second mechanism proposed by Slonczewski is biquadratic coupling mediated by paramagnetic atoms in the spacer layer [16]. These so-called "loose spins" can couple to both ferromagnetic layers via an indirect exchange, which also is responsible for $J_1$ (we will come back to this point later). The total interaction potential $U$ between loose spins and ferromagnetic layers is the vector sum of the interaction $U_1$ and $U_2$ of the loose spins with ferromagnetic layers 1 and 2, respectively, and can be expressed as

$$U(\theta) = [U_1^2 + U_2^2 + 2U_1U_2\cos(\theta)]^{1/2}, \qquad (2)$$

where $\theta$ is the angle between the two moments. The free energy per loose spin is

$$f(T,\theta) = -k_B T \ln\left(\frac{\sinh([1 + (2S)^{-1}]U(\theta)/k_B T)}{\sinh[U(\theta)/2Sk_B T]}\right), \qquad (3)$$

with $S$ the atomic spin and $T$ the temperature. The macroscopic free energy per unit spacer area $F = ca^{-2}f$, with $c$ the areal density of loose spins and $a$ the nearest neighbor distance between atoms, can be expanded in

$$F(\theta) = J_0 - J_1\cos(\theta) - J_2\cos^2(\theta) + \ldots, \qquad (4)$$

where

$$J_2(T) = -\frac{1}{2}ca^{-2}[f(T,0) + f(T,\pi) - 2f(T,\pi/2)] \qquad (5)$$

is the loose spins contribution to the biquadratic coupling. Previously, this loose spins model has been successfully

applied to explain the biquadratic coupling in the RKKY systems Fe/Al/Fe, Fe/Au/Fe, and Fe/Ag/Fe [16–19]. Especially in the Fe/Ag/Fe system the loose spins model has been proven to explain the biquadratic coupling directly by depositing an ultrathin Fe layer in the center of the Ag spacer. Without the ultrathin Fe layer this system shows no loose spins behavior.

Unlike the other mechanisms for biquadratic coupling, the loose spins model predicts a strong temperature dependence of $J_2$. Indeed, the huge increase of $J_2$ shown in Fig. 2(b) with decreasing temperature is a very strong indication for a loose spins origin. Figure 2(b) is supplemented with fits of $J_2(T)$ with the loose spins model (solid lines), assuming $S = 1$ and $U = U_1 = U_2$, which means that the loose spins are atoms near the midplane of the spacer or randomly distributed. The areal loose spins density $c$ and the interaction potential $U$ were adjusted for the fit. As can be seen in the figure, the loose spins model is in very good agreement with the experimental data. The density of loose spins following from the fits 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. In another approach one can assume $c = 2$ and allow for $U_1 \neq U_2$, which describes the case of two loose spins layers near the edge of the ferromagnetic interfaces [16]. However, with these assumptions our data cannot be described, because this leads to a plateau in $J_2$ for lower temperatures, not observed experimentally.

In the loose spins model the interaction potential $U$ is driven by the intrinsic bilinear coupling $J_1$ between the ferromagnetic layer and the loose spins in the spacer. Qualitatively this can be proven by measuring the thickness dependences of both the bilinear and the biquadratic coupling. Figure 3 shows $J_1$ and $J_2$ at room temperature as a function of the nominal Si spacer layer thickness measured on the wedge-shaped sample. We have plotted the coupling parameters only for $t_{Si}$ between 12.8 and 16.25 Å, when
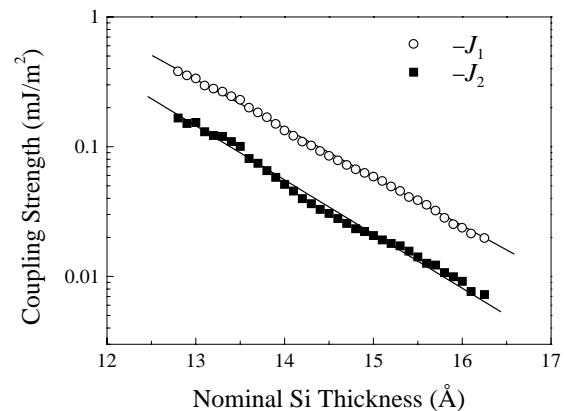


FIG. 3. Thickness dependence of $J_1$ (open circles) and $J_2$ (solid squares) at room temperature. The solid lines illustrate the exponential decay with the Si thickness.

$J_1$ and $J_2$ could be separated unambiguously. Indeed, both $J_1$ and $J_2$ decrease exponentially with the Si spacer layer thickness with approximately the same decay length. This shows that indeed both $J_1$ and $J_2$ are intimately related and find their origin in the same mechanism.

Furthermore, the unique exponential thickness dependence of the intrinsic exchange interaction enables us to perform a more quantitative analysis of $J_1$. From the fits for $J_2(t_{Si})$ we can obtain the strength of the driving intrinsic bilinear interaction $J_1$ under the assumption that the loose spins are located at the midplane. Given the exponential character of $J_1$ as shown in Fig. 3 and characterized by $\lambda$, we can calculate the intrinsic $J_1(t_{Si})$ at 0 K between the magnetic electrodes:

$$J_1(t_{Si}, 0\ \mathrm{K}) = a^{-2} e^{-t_{Si}/2\lambda} U. \qquad (6)$$

This results in $J_1 = 0.13,\ 0.10,\ 0.072,\ 0.058,\ 0.035,$ and $0.028\ \mathrm{mJ/m^2}$ for $t_{Si} = 14,\ 14.5,\ 15,\ 15.25,\ 16,$ and $16.25\ \text{Å}$, respectively, in good agreement with the actually measured low temperature bilinear coupling [see Fig. 2(a)]. This surprising result, given the simplicity of the model, leads to two important conclusions: First of all, the intrinsic $J_1$ is apparently the only contribution to the overall bilinear coupling and virtually no bilinear loose spins contribution is observed. Qualitatively this conclusion is also reflected in the relatively small temperature dependence of $J_1$ as already discussed earlier. Second, this is the first direct proof of one of the basic assumptions of Slonczewski's loose spins model that the interaction potential driving the coupling via loose spins is the same as the intrinsic interaction mechanism.

The absence of a bilinear loose spins contribution in systems where $J_2$ originates from loose spins is well documented and is not restricted to our rather unique Fe/Si system. Although the available data are scarce in literature, also in the RKKY-driven Fe/Ag/Fe system the bilinear loose spins contribution is only 20% of the value expected from the Slonczewski model [19]. Apparently, further refinements of the theory seem to be necessary. The prospect of predicting the bilinear loose spins contribution has been questioned already by Slonczewski because of scattering effects of the loose spins atoms on the electron waves neglected in the original model [20].

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