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Temperature Dependence of Interlayer Coupling in Fe/Si Superlattices

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Abstracts- **We have explored the temperature dependence of the interlayer coupling in Fe/$Fe_{1-x}Si_x$ superlattices ($0.5 \leq x \leq 1$). It is found that the Si content of the $Fe_{1-x}Si_x$ spacer greatly affects the temperature dependence of the bilinear and biquadratic coupling constants. Neither the "thickness fluctuation" model nor the "loose" spin model proposed by Slonczewski give satisfactory explanations to the temperature-dependent interlayer coupling. Instead, the present experimental results for all spacer compositions can be reproduced very well by the quantum interference model. We discuss the experimental results based on the above interlayer coupling models.**

Index terms — **Fe/Si(Fe) superlattice, interlayer coupling, remanence, biquadratic coupling**

I . INTRODUCTION

In spite of numerous studies on the interlayer coupling in Fe/Si superlattices,[1]-[4] the origin of the coupling is still an open question. There are several models to explain the coupling in the superlattices. First is the insulating spacer model in which hopping electrons in an amorphous Si spacer mediate the coupling.[1][5] Second is the semiconducting spacer model where thermally excited carriers in ε-FeSi or β-$FeSi_2$ contribute to the coupling.[4] Third is the metallic spacer model where conduction electrons in metallic silicides formed by interdiffusions at Fe/Si interfaces cause the coupling.[2][3] In order to clarify these controvertial problems, we recently investigated the coupling behaviors in Fe/$Fe_{1-x}Si_x$ ($0.4 \leq x \leq 1$) superlattices, where the Fe-Si alloy spacer changes from metallic to insulating with increasing the Si content.[6][7] Regardless of the spacer composition, all these superlattices exhibit similar coupling behaviors against the spacer thickness at room temperature. However, precise analyses of the bilinear and biquadratic coupling constants have revealed that the temperature dependence of the coupling constants varies sensitively with the spacer composition.[6][7]

In this article, we attempt to explain the temperature dependence of the interlayer coupling in Fe/$Fe_{1-x}Si_x$ superlattices based on the three kinds of coupling models; "thickness fluctuation"[8] and "loose spin"[9] models both proposed by Slonczewski and the quantum interference model.[10]

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II . EXPERIMENTAL PROCEDURE

Various Fe/$Fe_{1-x}Si_x$(x=0.54, 0.63, 0.73, 1.00) superlattices were grown on surface oxidized Si(100) substrates in a dc magnetron sputtering apparatus at ambient temperature. The superlattices were grown with the Fe layer thickness fixed at t_F=30Å and the nominal spacer thickness t_s varied from 3-70Å, with 22 bilayers. The details of the sample preparation conditions and characterizations are described elsewhere.[6][7]

III. RESULTS AND DISCUSSION

Figure 1 shows the remanence ratio for Fe(30 Å)/$Fe_{1-x}Si_x$(t_s) (x=0.54, 0.63, 0.73, 1.00) superlattices versus spacer film thickness t_s. We can notice that the interlayer coupling is initially ferromagnetic (F) and then oscillates from ferromagnetic to antiferromagnetic (AF) and goes toward non-coupling for all spacer compositions examined in the present experiment. Such changes in the coupling states against t_s were also confirmed by FMR measurements. Figure 2 showed the temperature dependence of the ramanence ratio for AF coupled samples. For the spacer with $x > 0.7$, M_r/M_s increases with decreasing temperature. In contrast, the ratio remains almost constant for x=0.63 over the whole temperature range of 100~480K. As pointed out by Fullerton et al.[11] and Kohlhepp et al.[12], the temperature dependence of M_r/M_s of Fe/Si superlattices could be understood by taking into account of a temperature-dependent biquadratic coupling term in addition to a bilinear one. As a possible origin of the biquadratic coupling in Fe/Si superlattices, they favor the two coupling models proposed by Slonczewski; one is the "thick-

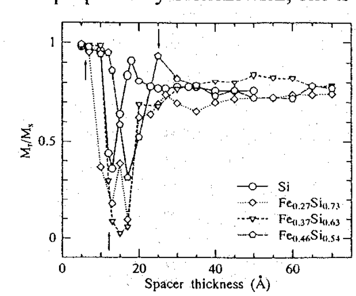

Fig. 1. Dependence of the remanence ratio (M_r/M_s) on the spacer thickness in Fe(30 Å)/$Fe_{1-x}Si_x$(t_s) (x=0.54, 0.63, 0.73, 1.00) superlattices.

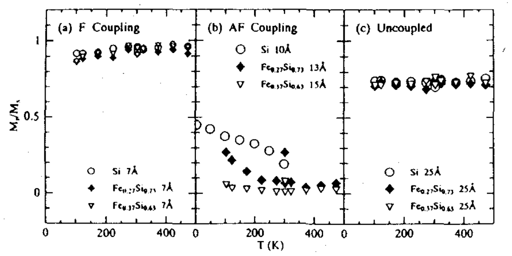

Fig. 2. Temperature dependence of the remanence ratio (M_r/M_s) in Fe(30 Å)/$Fe_{1-x}Si_x(t_s)$ (x=0.63, 0.73, 1.00) superlattices.

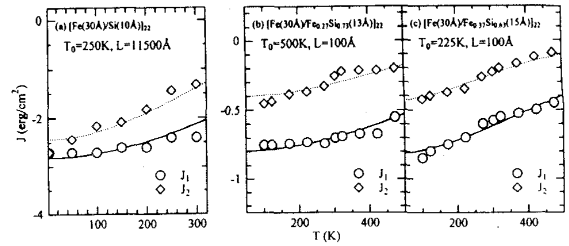

Fig. 3. Temperature dependence of the interlayer coupling constants(J_1, J_2) in the AF coupled Fe/$Fe_{1-x}Si_x$(x=0.63, 0.73, 1.00) superlattices fitted by "thickness fluctuation" model (the solid line : the calculated J_1, the dotted line : the calculated J_2).

ness fluctuation" model,[8] and the other is the "loose" spin model.[9] Based on these models, we first attempt to explain the temperature dependence of the interlayer coupling in Fe/$Fe_{1-x}Si_x$ superlattices.

The total energy of an exchange-coupled trilayer system is given by

$$E = -2M_s t_{Fe} H\cos\theta - J_1(T)\cos 2\theta - J_2(T)\cos^2 2\theta\,, \qquad (1)$$

where θ denotes the angle between the external field and the magnetization, $J_1(T)$ and $J_2(T)$ are the bilinear and the biquadratic coupling constants, respectively. All the magnetization curves measured in the present experiment can be fitted very well by this energy expression, thus we can evaluate the coupling constants for all spacer compositions. The coupling constants are plotted in Fig.3.

According to the quantum interference model by Bruno,[10] the bilinear coupling constant can be approximately expressed as

$$J_1(T)' = J_{10}'(T/T_0)/\sinh(T/T_0) \quad \text{for a metallic spacer}, \qquad (2)$$

and

$$J_1(T)'' = J_{10}''(T/T_0)/\sin(T/T_0) \quad \text{for an insulating spacer}, \qquad (3)$$

where J_{10}' and J_{10}" are the bilinear coupling coefficients, and T_0 is a quantity releavant to the wave vector and the spacer thickness. If the spacer is spatially inhomogeneous due to compositional fluctuations or interdiffusions,[6][11][12] the spacer properties will vary with position. Assuming for simplicity that the spacer is a mixture of metallic and insulating compounds, the general expression for the bilinear coupling is given by

$$(J_1)_{av} = \lambda J_1' + (1-\lambda)J_1''$$

$$= \lambda J_{10}'(T/T_0)/\sinh(T/T_0) + (1-\lambda)J_{10}''(T/T_0)/\sin(T/T_0)$$

$$(0 \leqq \lambda \leqq 1), \qquad (4)$$

where λ denotes the ratio of the metallic part to the total spacer.

By precise fitting of temperature dependence of the measured J_1 using Eq. (4) under the assumption of constant T_0, $\lambda J_{10}'$ and $(1-\lambda)J_{10}''$ were evaluated. According to the "thickness fluctuation" model,[8] a biquadratic coupling constant J_2 can be expressed as

$$J_2 = \left(4L(\Delta J)^2/\pi^3 A\right)\coth(\pi t_F/L)\,, \qquad (5)$$

where A is the exchange stiffness constant, L is the width of periodic terraces, and ΔJ is the spatial fluctuation of J_1 due to the roughness at the interface. On substituting Eqs.(2)-(4) into Eq.(5), the constant J_2 can be derived as;

$$J_2(T) = \left(4L/\pi^3 A\right)\left(\Delta J_{10}'\left[(T/T_0)/\sinh(T/T_0)\right]\right)^2 \coth(\pi t_F/L)$$

for a metallic spacer, (6)

$$J_2(T) = \left(4L/\pi^3 A\right)\left(\Delta J_{10}''\left[(T/T_0)/\sin(T/T_0)\right]\right)^2 \coth(\pi t_F/L)$$

for an insulating spacer, (7)

and

$$J_2(T) = \left(4L/\pi^3 A\right)\left(\Delta'(\lambda J_{10}')(T/T_0)/\sinh(T/T_0) + \Delta''((1-\lambda)J_{10}'')(T/T_0)/\sin(T/T_0)\right)^2 \coth(\pi t_F/L)$$

for a mixed spacer, (8)

here J_{10}' and J_{10}'' are the bilinear coupling constants at T=0, and Δ, Δ', and Δ'' stand for the coupling fluctuation due to the spacer thickness fluctuation.

Another model for the interlayer coupling is the "loose" spin model,[9] in which the interlayer coupling is mediated by the loose spins present within the spacer or adjacent to the interface. This model predicts the bilinear and biquadratic coupling constants as follows;

$$J_1 = \frac{k_B T}{2}\ln\left[\frac{\sinh(3T_0/T)\sinh(T_\pi/T)}{\sinh(3T_\pi/T)\sinh(T_0/T)}\right] \qquad (9)$$

and

$$J_2 = \frac{k_B T}{2}\ln\left[\frac{\sinh(3T_0/T)\sinh(3T_\pi/T)\sinh^2\left(\sqrt{T_0^2+T_\pi^2}/\sqrt{2}T\right)}{\sinh(T_0/T)\sinh(T_\pi/T)\sinh^2\left(3\sqrt{T_0^2+T_\pi^2}/\sqrt{2}T\right)}\right]$$

(10)

where $T_0 = (U_1 + U_2)/2Sk_B$ and $T_\pi = (U_1 - U_2)/2Sk_B$, and S is the local spin quantum number. Here U_1 and U_2 are conveniently dimensioned exchange-coupling fields due to the conduction-electron polarization fields induced by the two neighboring ferromagnetic layers. Since this model is based on the RKKY interaction, it is applicable only to the metallic spacer.

We found that the temperature dependence of J_1 in Fig.3 could be explained by either the quantum interference model (Eq.(4)) or the "loose" spin model (Eq.(9)) by choosing appropriate fitting parameters. As will be described later, however, the latter model failed in explaining the results for J_2. After the complete fitting of J_1, we performed fitting for J_2 according to the "thickness fluctuation" model in Eq.(8). The best fitted results are indicated by solid lines in Fig.3. It can be noticed that the fitting procedure is successful only for x=0.63, but not for x>0.7. Thus, the "thickness fluctuation" model can explain the coupling only for the $Fe_{0.37}Si_{0.63}$ spacer which is identified as a metallic conductor, but not for the spacer with x>0.7 which is in an intermediate state between metallic and insulating.[7] The "loose" spin model in Eqs. (9) and (10) also gave no satisfactory fitting results for the present experimetal data, as shown in Fig.4.

As mentioned above, it is hard to explain the interlayer coupling in the $Fe/Fe_{1-x}Si_x$ superlattices according to the two Slonczewski's coupling models. In contrast with these models, the quantum interference model[10] gives excellent fitting results to the present experimental data for both J_1 and J_2. As a higher order term of the interlayer coupling, the biquadratic coupling constant can be expressed as

$$(J_2)_{av.} = \lambda J'_{20}(2T/T_0)/\sinh(2T/T_0) + (1-\lambda)J''_{20}(2T/T_0)/\sin(2T/T_0) \qquad (11)$$

Fitting results by Eqs.(4) and (11) are shown in Fig.5. It was found that the fittings were very successful for all spacer compositions. Such good agreements between the experiments and the theory can be realized assuming that the $Fe_{1-x}Si_x$ spacer is purely metallic for x<0.7 and a mixture of metallic and insulating substances for $x \geqq 0.7$. These assumptions are supported by our recent measurements on the temperature

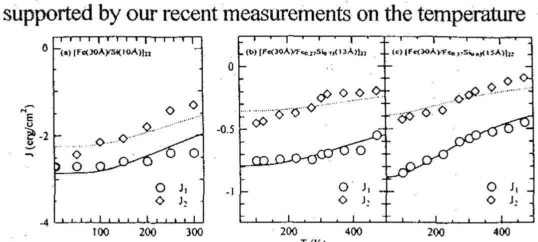

Fig. 4. Temperature dependence of the interlayer coupling constants(J_1, J_2) in the AF coupled $Fe/Fe_{1-x}Si_x$(x=0.63, 0.73, 1.00) superlattices fitted by "loose" spin model (the solid line : the calculated J_1, the dotted line : the calculated J_2).

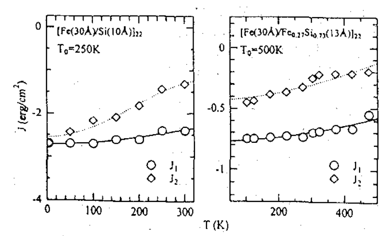

Fig. 5. Temperature dependence of the interlayer coupling constants(J_1, J_2) in the AF coupled $Fe/Fe_{1-x}Si_x$(0.73, 1.00) superlattices fitted by the quantum interference model (the solid line : the calculated J_1, the dotted line : the calculated J_2).

-dependence of the electric conductivity.[7]

In summary, we explored the interlayer coupling of $Fe/Fe_{1-x}Si_x$ (x=0.54, 0.63, 0.73, 1.00) superlattices, and found that the temperature dependence of the bilinear and biquadratic coupling constants were greatly influenced by the spacer composition. It was very difficult to explain the present experiments by either the "thickness fluctuation" model or the"loose" spin model proposed by Slonczewski. On the other hand, the quantum interference model gave satisfactory agree-ment between the experiments and the theory. We believe that the bilinear and biquadratic interlayer coupling found in $Fe/Fe_{1-x}Si_x$ superlattices are due to the intrinsic quantum interference effect.

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