Temperature dependence of the resonant exchange coupling between two ferromagnets separated by a nonmetallic spacer

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By considering the quantum-size effect arising from ferromagnets (FM) thickness and the interference of tunneling electrons in an FM layer, we have investigated the bilinear coupling (J_1) and the intrinsic biquadratic coupling (J_2) between two FM's separated by a nonmetallic spacer. It is found that both J_1 and J_2 are decreasing oscillatory functions of FM thickness with many sharp resonant peaks. Near these peaks, the strengths of J_1 and J_2 have negative temperature coefficients. In striking contrast, in the region of nonresonant coupling, the thermal excitations of conduction electrons tend to enhance the antiferromagnetic coupling, but suppress the FM one, and make J_2 oscillate in sign with the increasing temperature. In addition, we also find that a nonmonotonous temperature coefficient of J_1 can be derived by lowering the barrier height to some extent. The calculation about the density of states (DOS) indicates that these different thermal features are related to the relative magnitudes of DOS above Fermi surface between the parallel and antiparallel alignments of FM's. Therefore the present paper provides a feasible explanation for the recent experimental observations.

I. INTRODUCTION

The discoveries of weak antiferromagnetic (AF) interlayer exchange coupling across amorphous (α)Si in Fe/α-Si/Fe sandwiches[1] and strong AF coupling in Fe/Si multilayers[2] have initiated considerable interest in magnetic multilayers with nonmetallic spacers.[3–22] Especially, the recent attention is paid to the temperature dependence of exchange coupling in Fe/Si(FeSi,Ge) heterostructures. It was observed that, in Fe/α-Si(Ge)/Fe trilayers prepared at low temperature, the strength of the weak AF coupling had a positive temperature coefficient (heat-induced coupling).[1,3,4] In striking contrast, in the multilayers of Fe/Si(Ge) prepared at room temperature (RT), the AF coupling exhibited a negative temperature coefficient.[2,6–12] Moreover, a nonmonotonous temperature dependence of J_1 was recently found in Fe/α-SeZn/Fe sandwich,[5] where the ferromagnetic (FM) coupling can reversibly transform to AF upon heating. Walser and co-workers[3–5] argued that the positive temperature coefficient for the AF coupling is due to the thermal excitation of localized defect states in the gap of the spacer. While the negative one was attributed to the fact that spacers of Fe/Si multilayer prepared at RT are not amorphous semiconductors, but crystalline metallic Fe-Si compounds. [2,7] So this kind of Fe/Si multilayers has the ''full'' metallic structure, and it is natural the AF coupling has the conventional negative temperature coefficient. Furthermore, based on the analyses of hysteresis loops, Fullerton and Bader suggested that a strongly temperature-dependent biquadratic coupling (J_2) is responsible for the observed remanent magnetization at low temperature.[10,13,14] However, it was refuted by Kohlhepp that the vertical and lateral variations in bilinear AF and FM coupling can ''mimic'' a strong biquadratic coupling.[15,16] Therefore the nature of exchange couplings J_1 and J_2 in Fe/Si(Ge) multilayers is still unclear and is a matter of controversial discussion. In fact, such a biquadratic coupling was originally discovered by domain microscopy in Fe/Cr/Fe trilayers, where J_2 was ascribed to thickness fluctuation mechanism proposed by Slonczewski.[17]

Several theoretical models have been proposed to account for the behavior of interlayer exchange coupling across the nonmetallic spacer. The first is the spin-current model of Slonczewski at zero temperature,[18] the essence of which is to construct the stationary wave functions from the Schröd inger equation and the continuous boundary conditions of the wave functions. The coupling can be derived from the torque produced by rotation of the magnetization of one FM relative to that of the other. The second is the quantum interferences model of Bruno,[19] which ascribes the coupling to the interferences of electron waves in the barrier layer due to the spin-dependent reflections at the interfaces. The coupling can be expressed in terms of the spin asymmetry of the reflection. It succeeds in obtaining the increasing AF coupling with temperature. At $T=0$, it reduces to the Slonczewski's spin current model. In addition, there are the Kondo lattice model of Shi, Singh, and Klein[20] and the nonequilibrium Keldysh formalism.[21,22] It is noted that the negative temperature coefficients for the strengths of J_1 and J_2 cannot be derived from above mechanisms.

As far as above models themselves are concerned, they all took the approximation of semi-infinite FM layer. In fact, the FM samples used in the experiments are not thick enough, but usually in nm order, which is less than the electrons mean free path. So the quantum-size effect must be remarkable and dominant in the tunneling process. Furthermore, spin-dependent reflection from the FM surfaces and interfaces could give rise to quantum confinement in FM layers, setting up spin-dependent quantum-well states. Thus the models with semi-infinite FM's have neglected the contribution of quantum-well states, and need to be improved.

In present work, with emphasis on the quantum-size effect arisen from FM thickness, we extend the quantum-well model[23–25] of metallic multilayer to the case of nonmetallic

spacer. The interfaces of FM/$I(S)$ make up the double quantum-well with two FM surfaces. Owing to the tunneling effect and the reflections of electron waves from the surfaces of FM, the conduction electrons would interference within the FM wells and give rise to the resonant exchange coupling at certain condition. On the whole, if the metallic layers are nonmagnetic metals, we could not observe any net ''current'' between two metallic layers in the case of equilibrium with zero bias. When the metallic layer is FM metals, in the right FM layer, the localized spins would interact with the polarized conduction spin density arising from the localized spin on the left side; whereas in the left FM layer, the localized spin interacts with the polarized conduction spin density arising from the localized spin on the right side. The total exchange energy for spins on different sides of the barrier is the sum of these two contributions. So there still exists a net spin current[18,26] even in the zero bias case. It is just this spin current that make the two FM's couple each other.

Our numerical results indicate that: the interlayer exchange coupling (J_1 and J_2) oscillate with the variation of FM thickness and molecular filed. During oscillation, both J_1 and J_2 have two kinds of amplitudes: one is resonant exchange coupling with some very sharp peaks; the other is the nonresonant coupling with small magnitude but large width. Most importantly, the resonant coupling strength has a negative temperature coefficient. On the other hand, for the nonresonant coupling, the thermal excitation of conduction electrons tends to enhance the AF coupling, but suppress the FM one. Furthermore, a nonmonotonous temperature dependence of J_1 can be derived by lowering the barrier height. Therefore all of the observed phenomena about the thermal dependences of exchange couplings can be explained qualitatively within present work.

Similar to the treatments by Edwards *et al.*[25] and Bruno and Chappert,[27] in calculating the temperature dependence of exchange coupling, we only consider the contribution from Fermi-Dirac distribution to the system total energy, neglecting the influence of thermal variation of magnetization in FM's.

The rest of this paper is organized as follows. In Sec. II, we introduce briefly the double quantum-well model and the main procedure to calculate the bilinear and biquadratic coupling at nonzero temperature. The numerical results and discussion are displayed in Sec. III, and a brief summary is given in Sec. IV.

II. DOUBLE QUANTUM-WELL MODEL

Consider a sandwiched structure $FM_1/I(S)/FM_2$, where $I(S)$ denotes the barrier layer with thickness d_b. For the typical FM like Fe, Ni, and Co, due to the relative narrow $3d$ subbands, we assume the direct contribution from d electrons to the tunneling across the barrier can be neglected. Although this is a gross simplification, it agrees with the band-structure calculation of Tsymbal and Pettifor.[28] So the exchange coupling between two FM's originates from the interaction of conduction electrons with the system of uncompensated magnetic moments of d electrons. This lead us to the approximation that neglect the direct exchange interaction between d electrons and only takes the s-d exchange interaction into account. So the width of $3d$ subbands could be chosen as zero, corresponding to the complete localization.

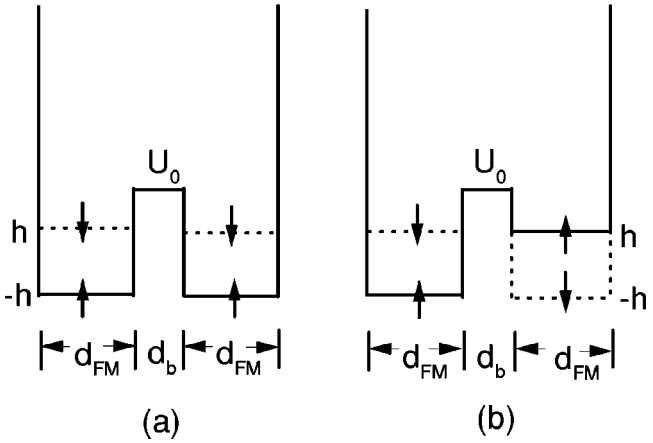

FIG. 1. Schematic illustration of the double quantum-well model. The spin-dependent effective potential in (a) and (b) correspond to the parallel and antiparallel configurations of two FM's. The solid and dotted lines correspond to majority spin electrons and minority spin ones, respectively.

When the ferromagnetism is very strong, we can neglect the fluctuations of localized spin density, and take the mean-field approximation to the s-d exchange interaction, as long as the FM layer is not too thin. But speaking strictly, the fluctuation of localized spin density is closely related with the magnetic order of FM, so the mean-field approximation would lead to an ''enhanced'' magnetic order and an enhanced spin polarization of s electrons. However, if we only consider the thick FM layer with strong ferromagnetism, such as Fe, Co, and Ni, the influence of spin fluctuation to the s-d exchange may be not important. So the mean-field approximation may be a effective method to describe the s-d exchange interaction, if we do not involve the critical behavior nearby the Curie temperature of FM's.

For simplicity, we assume FM_1 and FM_2 have the same thickness d_{FM} and the same magnitude of molecular (mean) field h, but different magnetization directions, which makes $\pm\theta/2$ with the z axis, respectively (the film plane is parallel to y-z plane, and perpendicular to the x axis). Within the spin polarized mean-field approximation to s-d exchange interaction, the conduction electrons move freely in the film plane, but are subject to a rectangular barrier U_0 and two infinitely high barriers in the x direction. The spin-dependent effective potential for conduction electrons is shown in Fig. 1, where the barrier makes up the double quantum well with the two FM's. The relevant eigenenergies can be determined from the Schrödinger equation with single-electron Hamiltonian

$$H=-\nabla^2+U(x)+\mathbf{h}(x)\cdot\sigma, \tag{1}$$

where σ is Pauli spin operator, $U(x)$ represents the potential profile and $\mathbf{h}(x)$ the mean-field of s-d exchange coupling (molecular field):

$$U(x)+\mathbf{h}(x)\cdot\sigma=\begin{cases} h[\cos(\theta/2)\sigma_z-\sin(\theta/2)\sigma_y], & \mathrm{FM}_1 \\ U_0, & I(S) \\ h[\cos(\theta/2)\sigma_z+\sin(\theta/2)\sigma_y], & \mathrm{FM}_2 . \end{cases} \tag{2}$$

By means of the rotation operator D,

$$D=\cos\left(\frac{\theta}{4}\right)-i\mathbf{e}_x\cdot\sigma\sin\left(\frac{\theta}{4}\right)\begin{bmatrix}\cos\left(\frac{\theta}{4}\right) & -i\sin\left(\frac{\theta}{4}\right)\\ -i\sin\left(\frac{\theta}{4}\right) & \cos\left(\frac{\theta}{4}\right)\end{bmatrix}, \tag{3}$$

we can take the unitary transformation to the Hamiltonian in FM layer with $H'=D^{-1}HD$, so the eigenfunctions can be derived by $\Psi=D\Psi'$.

As the structure has the rotational symmetry of π angle round the z axis, we can use the parity operator $T=I\sigma_z$ to simplify the calculation. By means of the boundary conditions of wave functions and their first derivatives, we can derive strictly the eigenenergy spectrum E_{xj} from[29]

$$\frac{AD+BC}{AD-BC}=\pm\cos(\theta/2), \tag{4}$$

where $\theta=0,\pi$ and $\pi/2$ correspond to the parallel, antiparallel, and 90° configuration of two FM's, $\pm$ the eigenvalues of the parity operator T, and

$$\begin{aligned}A&=p\cos(\alpha)\sinh(\gamma)+r\sinh(\alpha)\cosh(\gamma),\\ B&=p\cos(\alpha)\cosh(\gamma)+r\sinh(\alpha)\sinh(\gamma),\\ C&=q\cos(\beta)\sinh(\gamma)+r\sin(\beta)\cosh(\gamma),\\ D&=q\cos(\beta)\cosh(\gamma)+r\sin(\beta)\sinh(\gamma),\end{aligned} \tag{5}$$

with $\alpha=pd_{\mathrm{FM}}$, $\beta=qd_{\mathrm{FM}}$, $\gamma=rd_b/2$; $p=(E_x-h)^{1/2}$, $q=(E_x+h)^{1/2}$, $r=(U_0-E_x)^{1/2}$.

Based on the derived eigenenergies spectrum, and taking into account the variation of Fermi-Dirac distribution for conduction electrons, we can calculate the total energy of the system at any temperature, which can be expanded in powers of $\cos(\theta)$ as

$$E(\theta,T)=E_0(T)+SJ_1(T)\cos(\theta)+SJ_2(T)\cos^2(\theta)+\cdots, \tag{6}$$

where J_1 and J_2 are the bilinear and biquadratic coupling constants, S denotes the film area. For $J_1>0$, the exchange coupling favors AF coupling; whereas $J_2>0$ favors the 90° alignment. Neglecting the higher-order coefficients in the expansion, the coupling parameters can be determined from

$$J_1(T)=\frac{1}{2S}[E(0,T)-E(\pi,T)] \tag{7}$$

and

$$J_2(T)=\frac{1}{2S}[E(0,T)+E(\pi,T)-2E(\pi/2,T)] \tag{8}$$

with

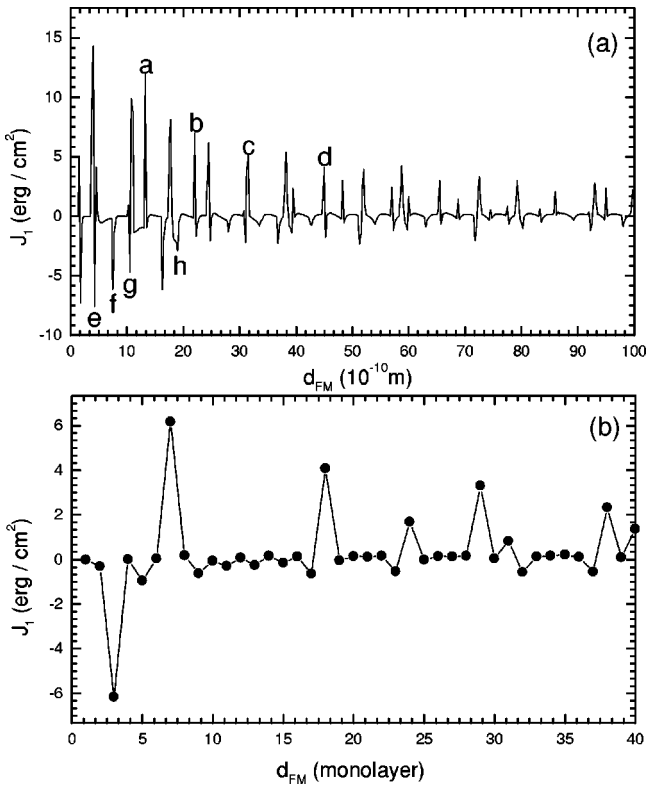

FIG. 2. Dependence of bilinear coupling J_1 on the continuous FM thickness (a) and on the discrete FM thickness in unit of 0.25 nm (b), where the relative barrier height $U-E_F=0.025$ eV, $h=1.8$ eV, $E_F=2.6$ eV, $d_b=1.0$ nm, and $T=0$.

$$E(\theta,T)=\frac{S}{4\pi^2}\sum_{j=0}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\times\frac{E_{xj}(\theta)+k_y^2+k_z^2}{\exp\{[E_{xj}(\theta)+k_y^2+k_z^2-E_F]/k_BT\}+1}dk_ydk_z. \tag{9}$$

It should be noted that, in some references,[13,14,16,26] the ''+'' in Eq. (6) was replaced by ''−.'' So the coupling parameters J_1 and J_2 have the opposite meaning: $J_1>0$ correspond to FM coupling, $J_2<0$ favors the 90° alignment of two adjacent FM's.

III. CALCULATING RESULTS

Based on the formulas (6)–(8), we can calculate the exchange coupling J_1 and J_2 at any temperature. Now we take Fe/ε-FeSi/Fe sandwich as a concrete example to investigate the behavior of exchange couplings. The Fermi energy and molecular field for Fe is derived from the bandwidth[30] and polarization measurement near Fermi surface:[31] $E_F=2.6$ eV, $h=1.8$ eV, the barrier height for the semiconductor spacer ε-FeSi (Ref. 6) is taken as: $U-E_F=0.025$ eV.

A. FM thickness dependences of exchange coupling J_1 and J_2

First of all, let us investigate the dependences of J_1 and J_2 on FM thickness at $T=0$, which are shown in Figs. 2 and 3,

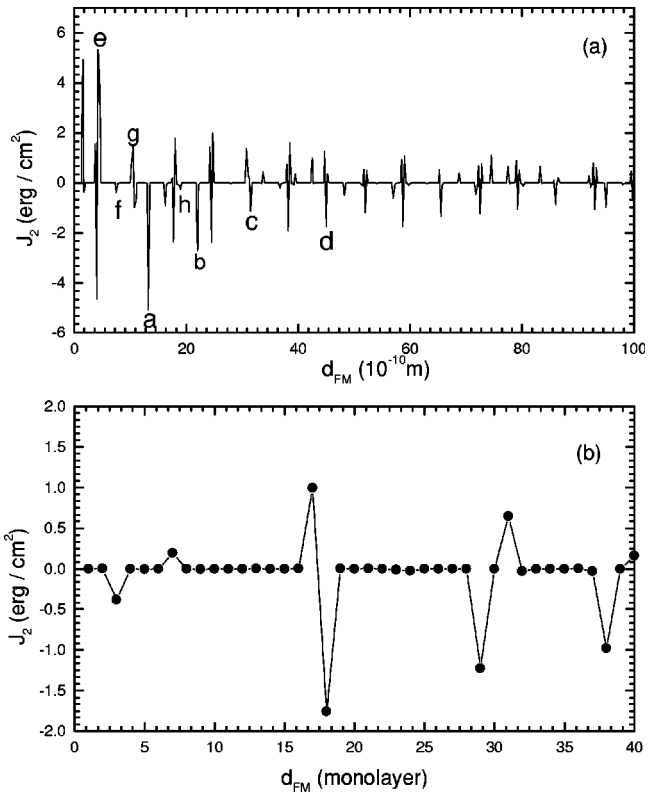

FIG. 3. Dependence of biquadratic coupling J_2 on the continuous FM thickness (a) and on the discrete FM thickness in unit of 0.25 nm (b), where $U-E_F$, h, E_F, d_b, and T are the same as those in Fig. 2.

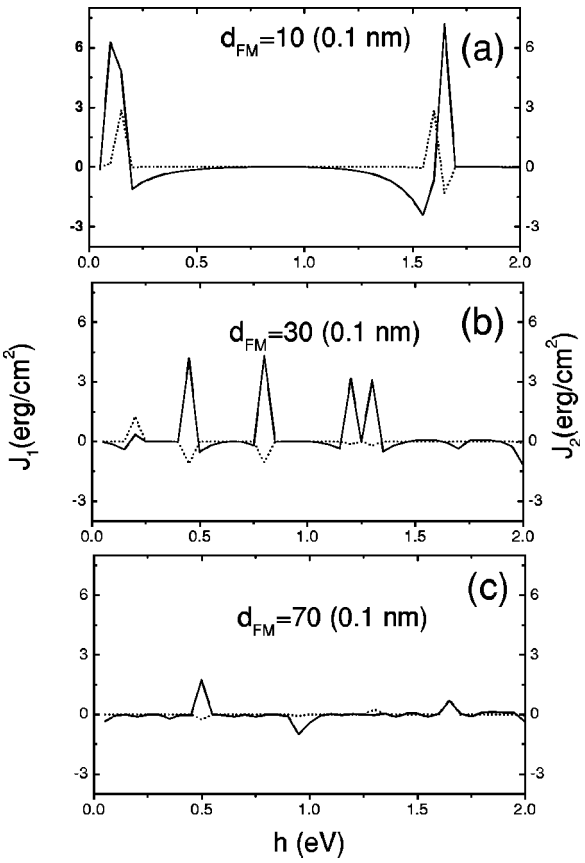

FIG. 4. Dependences of exchange coupling J_1(solid) and J_2 (dotted) on molecular field h, where d_{FM} is shown in (a), (b), and (c), $U-E_F$, E_F, d_b, and T are the same as those in Fig. 2.

respectively. It can be seen that, from the top panels of the two figures, both J_1 and J_2 are decreasing oscillatory functions of the continuous FM thicknesses. During oscillation, they exhibit many sharp resonant peaks with the almost same oscillatory phase. Furthermore, the peak amplitudes are much stronger than the nonresonant coupling. So the exchange coupling could be classified into two kinds: one is the resonant coupling with a large magnitude but a narrow width; the other is the nonresonant coupling with a large width but a very small magnitude. In the experimental observation, it is more easily to measure the weak nonresonant coupling for its large width.

In order to compare our results with experiments more realistically, in the bottom panels of Figs. 1 and 2, we have plotted dependences of J_1 and J_2 on the discrete FM thickness in unit of 0.25 nm (thickness of an atomic monolayer). Compared to results in the top panels, one finds that the oscillation gets ''aliased'' and the coupling strengths decrease greatly, due to the sharpness of resonant peaks for the continuous FM thickness. Moreover, it is also found that from Fig. 3, J_2 is almost zero except a few resonant peaks. This is the reason why the large biquadratic coupling is seldom observed in experimental measurements in Fe/Si system.

As for the mechanism of the resonant exchange coupling, it can be attributed to the quantum-size effect and the interference of tunneling electronic in the FM well. In general, the film thickness is in order of nm, which is less than the electrons mean free path. The quantum-size effect must be remarkable. Furthermore, due to the confinement of the FM surfaces with a infinite barrier height, the tunneling electrons subject the spin-dependent reflection from surfaces and interfaces, so they could interference in the FM wells, setting up the spin-dependent quantum-well states. At certain condition, this kind of interference becomes maximum, so the resonant exchange coupling takes place.

B. Molecular-field dependences of exchange coupling J_1 and J_2

As can be imagined, the interference of tunneling electrons should also be mediated by the relative depth between two quantum wells, represented by the molecular field h. So we plot the dependences of exchange couplings on molecular field h at zero temperature in Fig. 4. It is observed that both J_1 and J_2 oscillates with the increasing molecular field. During oscillation, the region of AF coupling is much smaller than that of FM one. In other words, most values of h favor the FM coupling. Therefore we can conclude that it is not always that the larger h is, the stronger exchange coupling becomes. Moreover, comparing Figs. 4(a)–(c), one can also find the oscillation amplitude decreases, but the frequency increases with the increasing FM thickness, this is in agreement with the conclusion in Figs. 2 and 3.

C. Temperature dependences of exchange coupling

In this subsection, we study the temperature dependences of bilinear coupling J_1 and biquadratic coupling J_2. In the

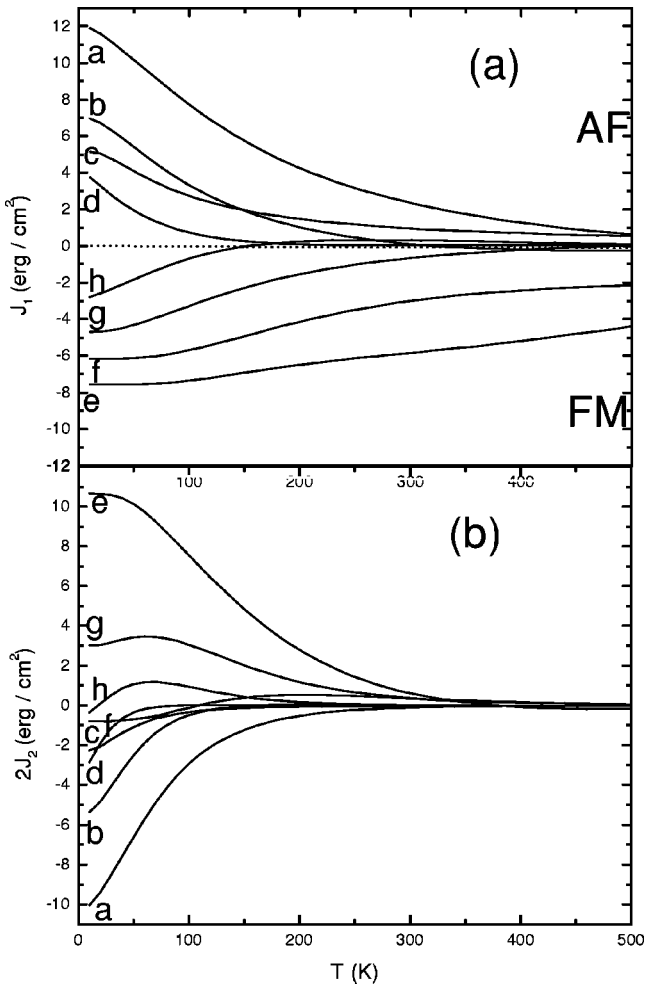

FIG. 5. Resonant exchange couplings J_1 (a) and $2J_2$ (b) vs temperature for a variety of FM thicknesses. Curves a, b, c, d, e, f, g, and h correspond to the resonant peaks represented by the same letters in Figs. 2 and 3, $U-E_F$, E_F, h, and d_b are the same as those in Fig. 2.

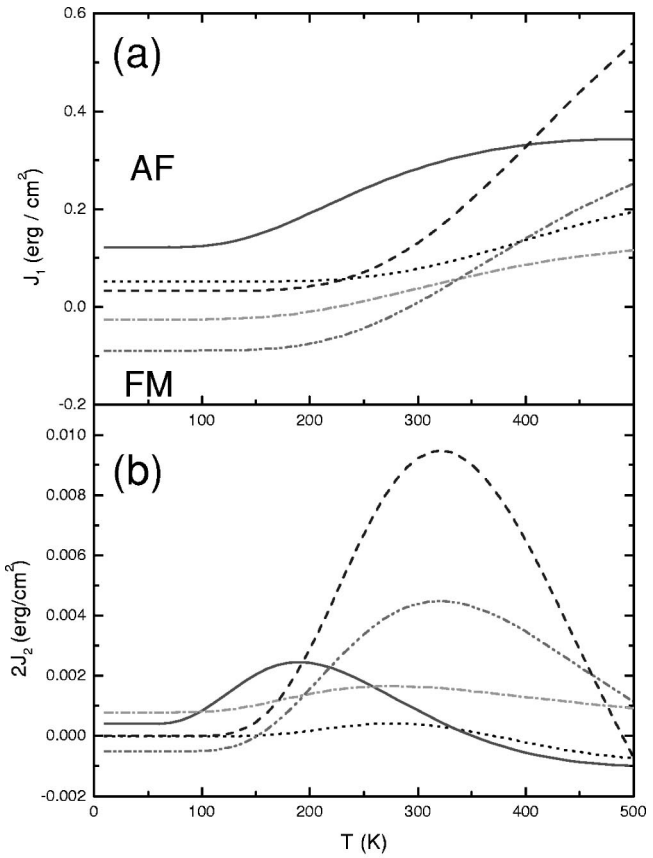

FIG. 6. Nonresonant exchange couplings J_1 (a) and $2J_2$ (b) vs temperature for a variety of FM thicknesses. The solid, dashed, dotted, dash-dotted, and dash-dot-dotted curves correspond to FM thickness 2.6, 0.9, 1.5, 2.65, and 2.85 nm, respectively. $U-E_F$, E_F, h, and d_b are the same as those in Fig. 2.

experimental observation performed by Fullerton and Bader[10] and by Kohlhepp *et al.*,[16] the saturation field H_S was used to fit the hysteresis loops with the relation $H_S=4(J_1+2J_2)/M_S t_{\mathrm{Fe}}$, where M_S is the magnetization and t_{Fe} is the FM thickness. Thus they used $J_1(T)$ and $2J_2(T)$ to characterize the thermal behaviors of exchange couplings. Moreover, as pointed out by Erickson, Hathaway, and Cullen,[26] when $J_2<0$ and $2J_2<-|J_1|$, the exchange coupling is noncolinear, and the canting angle is determined by $\cos\theta=J_1/|2J_2|$. Particularly, when $J_1\rightarrow 0$, 90° coupling takes place. Therefore from J_1 and $2J_2$, we can obtain the information of magnetic configuration for the adjacent FM layers. Hereafter, we also use $2J_2$ to characterize the thermal behavior of biquadratic coupling.

1. Resonant exchange coupling

The temperature dependences of resonant J_1 and $2J_2$ are shown in Fig. 5, where the curves a, b, c, d, e, f, g, and h correspond to the resonant peaks in Figs. 2 and 3. It is found that both the strengths of J_1 and $2J_2$, except the curve ''h,'' decrease exponentially with the increasing temperature, whether they are positive or negative. The amplitude of $2J_2$, being comparable with J_1, decrease more rapidly than J_1 does. This result is in agreement with the phenomena observed in Fe/Si multilayers,[6–12] where the biquadratic coupling was found to be more temperature dependent than bilinear coupling is. Thus we can make an inference that the experimental observed negative temperature coefficient for AF coupling and the strong biquadratic coupling term is possibly due to the resonant exchange interaction.

In order to explore the microscopic origin of above phenomenon, we have calculated the densities of states (DOS) for the parallel and antiparallel configurations of FM's, represented by DOS(0) and DOS(π), respectively. When the coupling is AF, $E(0,T)-E(\pi,T)$ is positive, we find that DOS(0) is smaller than DOS(π) above the Fermi surface, so $E(0,T)-E(\pi,T)$ decreases with increasing temperature, and the strength of J_1 decreases correspondingly. When the coupling is FM, $E(0,T)-E(\pi,T)$ is negative, we find that DOS(0) is larger than DOS(π) above the Fermi surface, so $|E(0,T)-E(\pi,T)|$ decreases with increasing temperature, and the strength of J_1 decreases correspondingly.

2. Nonresonant exchange coupling

As for the nonresonant coupling, it can be seen from Fig. 6 that, with the increasing temperature, the positive J_1 increases, but the negative J_1 also increases until zero. At certain condition, J_1 could change from positive value to nega-

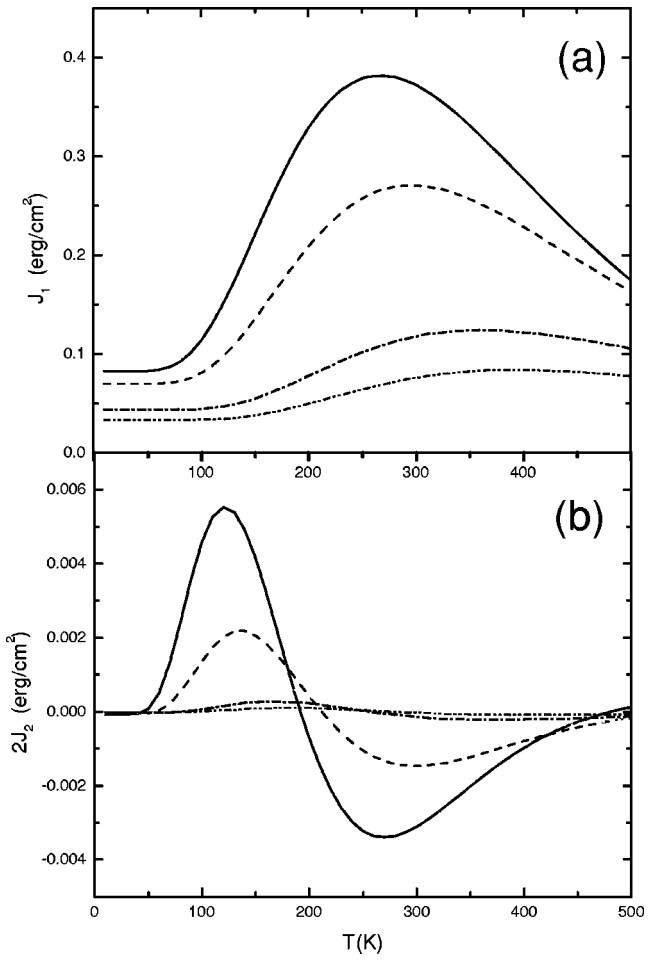

FIG. 7. Influence of barrier height on the temperature dependences of the nonresonant coupling J_1 (a) and $2J_2$ (b), where the solid, dashed, dotted, dash-dotted, and dash-dot-dotted curves correspond to the potential height $U-E_F=0.025$, 0.06, 0.1, 0.15, and 0.2 eV, respectively, $d_{\mathrm{FM}}=3.0$ nm, E_F, h, and d_b are the same as those in Fig. 2.

tive one. In other words, in the case of nonresonance, the strength of AF coupling has a positive temperature coefficient, but the strength of FM coupling has a negative one. This means that the thermal excitation of conduction electrons tends to enhance the AF coupling, but suppress the FM one. This result corresponds to the phenomena observed in Fe/α-SeZn/Fe sandwiches,[5] where the FM coupling could be converted to AF coupling upon heating. Furthermore, one can find that the magnitude of $2J_2$, compared with J_1, is so small such that it could be neglected. Therefore, when the exchange coupling is not resonant, J_2 is very difficult to be observed for its weakness.

In the same principle, these temperature-dependent features can be also traced to the behaviors of DOS above Fermi surface. It is calculated that, whether J_1 is positive or negative, DOS(0) is always larger than DOS(π). So the positive J_1 increases, and the strength of negative J_1 decreases with increasing temperature.

In addition, we have also studied the influence of barrier height on the temperature dependences of nonresonant couplings, which is displayed in Fig. 7. With the temperature changes from 0 to 500 K, J_1 increases monotonously for the high barrier, such as the dash-dot-dotted line with $U-E_F=0.2$eV; J_1 varies nonmonotonously from increasing to decreasing for the low barrier, such as the solid, dashed, and dotted lines. Meanwhile, $2J_2$ varies with the temperature more rapidly than J_1 does. Between 0 to 500 K, the biquadratic coupling could oscillate in sign under any barrier height. Therefore the barrier height could also mediate the monotonousness of the temperature dependence of bilinear coupling J_1.

Calculation about the DOS indicates that: for the AF coupling with a nonmonotonous T dependence, we find that DOS(0) $-$ DOS(π) changes from positive to negative value, with the energy increasing from Fermi surface. Hence, with the increasing temperature, $E(0,T)-E(\pi,T)$ has a transition point, before which J_1 has a positive temperature coefficient, and after which it has a negative one. Therefore we can make an inference that, at the transition temperature, there exists a equilibrium between two factors: one is the contribution from the electrons below the barrier, which favors the positive temperature coefficient for the strength of J_1; the other is that from the electrons above the barrier height, which favor the negative one, just like the conduction electrons in the metallic multilayers.

IV. SUMMARY

In this paper, based on the double quantum-well model, we have studied the temperature dependences of the interlayer exchange coupling between two FM's separated by a nonmetallic spacer. It is found that J_1 and intrinsic J_2 oscillate with the variation of FM thickness and molecular field. During oscillation, both the couplings exhibit some sharp resonant peaks. Near the resonant peaks, for the AF (FM) coupling, due to the fact that DOS(0) is smaller (larger) than DOS(π) above Fermi surface, the strength of J_1 has a negative temperature coefficient, wether the coupling is AF or FM. For the nonresonant coupling, as the DOS(0) is always larger than DOS(π), so AF coupling increases, and the strength of FM coupling decreases with the increasing temperature. Moreover, a nonmonotonous temperature coefficient of J_1 can be derived by lowering the barrier height to some extent. Therefore the above findings provide a qualitative explanation for the recent experimental observations.

However, the present model, being total-energy calculation like, does not involve the accurate description of temperature dependence of the magnetization in FM's. Furthermore, it does not take into account the thickness fluctuations, which will become significant when the films get very thin. It is therefore not possible to make a detailed comparison between the experimental observation and our calculating results. However, the present work does provide a initial qualitative appraisal for the temperature dependences of the resonant and nonresonant exchange couplings in the Fe/FeSi system, particularly, for the interference of tunneling electrons in the double quantum-well.

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