Interlayer exchange coupling mediated by a nonmagnetic spacer layer with a large electron-electron exchange interaction

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We derive an expression for the exchange coupling between two ferromagnetic layers separated by a nonmagnetic spacer metal having large exchange enhancement in the paramagnetic susceptibility. The theory of the exchange coupling between two magnetic spins embedded in a nonmagnetic metal, in which the random-phase approximation was used for the electron-electron exchange interaction, is extended to the exchange coupling between ferromagnetic layers in magnetic superlattices. The exchange coupling decreases with an increasing interlayer thickness in a nontrivial manner. An oscillatory behavior superimposed on the exponentially decreasing background is clearly shown. The effect due to the electron-electron exchange interaction in the nonmagnetic spacer metal produces a ferromagnetic bias to the oscillatory exchange coupling for low spacer thicknesses. The regions of the antiferromagnetic exchange coupling may disappear because of the ferromagnetic bias depending on the degree of the exchange enhancement. However, the traces of the disappeared regions can be recognized as dips or easy slopes in the region of the ferromagnetic exchange coupling. It is shown from the numerical results that in the case of magnetic superlattices with Pd interlayers, the first and second antiferromagnetic exchange couplings disappear and the third antiferromagnetic one appears, and the theoretical result agrees well with the experimental result. The ferromagnetic bias is less expected for the Pt interlayer than for the Pd interlayer because of a relatively weak electron-electron exchange interaction. [S0163-1829(97)04337-3]

I. INTRODUCTION

Since Grünberg *et al.*[1] discovered that the magnetic moment in Fe layers separated by a spacer Cr layer in Fe/Cr/Fe sandwiches could be coupled to each other antiferromagnetically, and Baibich *et al.*[2] studied the giant magnetoresistance (GMR) in Fe/Cr superlattices resulting from the interplay between electron transport and magnetic behavior, a number of studies have been made of these phenomena. Parkin, More, and Roche[3] discovered that the exchange coupling could oscillate between ferromagnetic (FM) and antiferromagnetic (AFM) coupling as a function of nonmagnetic (NM) spacer thickness. Parkin[4] also revealed that the oscillatory exchange coupling between two FM layers separated by ordinary NM transition metals or noble metals is quite a common phenomenon.

There has been a great interest in GMR, not only for the underlying mechanism, but also for its practical device applications such as magnetoresistive heads and sensors. Low saturation magnetic fields resulting from a low AFM coupling strength are desirable for the enhancement of the sensitivity of device applications. The technology for the enhancement has been studied in methods such as adopting magnetic materials as soft as possible for FM layers,[5] choosing a thicker NM spacer layer corresponding to the second peak in the GMR effect,[6] employing a spin-valve structure with an uncoupled behavior at large NM spacer layer thicknesses,[7] and inserting subsidiary soft FM layers.[8]

There have been many theoretical investigations to attain some better insight into the mechanism of the oscillatory exchange coupling. The spin polarization of conduction electrons in the NM spacer via the Rudermann-Kittel-Kasuya-Yoshida (RKKY) interaction and the discreteness of the spacer thickness due to a stack of atomic monolayers (ML), known as aliasing, were taken into consideration in order to explain the oscillatory exchange coupling.[9,10] Very recently, the effect of a composite NM layer which consists of a conventional NM layer and a potential scattering layer inserted in it was analyzed, and it was found that the transition from FM to AFM coupling or the reverse could be induced by changing the potential parameters under a definite structure of the superlattices.[11] The Anderson model of the s-d mixing at the interfaces between FM and NM layers was applied to the problem of the exchange coupling in magnetic superlattices, and two types of couplings, i.e., RKKY-like and superexchange interactions, were shown.[12] A spin-dependent quantum well description of the electronic structure has also been proposed for the coupling mechanism.[13] The quantum interference due to the spin-dependent reflections of Bloch waves at the interfaces between NM and FM layers has been considered as a general approach to the problem of the exchange coupling.[14] This approach is an analogy of an optical Fabry-Pérot resonator and is closely related to the quantum well description.

The exchange effect due to the electron-electron (e-e) Coulomb interaction among d electrons in the NM spacer layer has not been considered so far in the context of the exchange coupling theory in magnetic superlattices. However, since the e-e exchange interaction is strong in Pd and Pt metals, it should be taken into consideration in the case of those NM spacer layers.

Especially, Pd is a more interesting metal magnetically since it is near the critical boundary for ferromagnetism. It is a nonmagnetic metal having large exchange enhancement in the paramagnetic susceptibility, and dilute Fe-Pd and Co-Pd alloys show giant magnetic moments as large as $10\mu_B$ per impurity associated with impurity atoms. This was first observed through the paramagnetic susceptibility and the ferromagnetic saturation magnetization.[15]

The giant magnetic moments of Fe or Co atoms in those alloys indicate a long-range ferromagnetic spin coupling associated with a long-range spin polarization in Pd host metal. Therefore, it has been considered that the giant magnetic moments in Pd alloys are certainly related to the large exchange enhancement in the paramagnetic susceptibility of Pd metal.

The same phenomenon of spin polarization has been observed in Fe/Pd (Refs. 16–18) and Co/Pd (Refs. 16, 19 and 20) superlattices, in which the ferromagnetic polarization of Pd atoms at the interfaces was found. Gelinski *et al.*[17] observed magnetic properties of the Fe/Pd/Fe trilayers and individual Fe/Pd and Pd/Fe bilayers by using ferromagnetic resonance (FMR) and Brillouin light scattering (BLS). The samples were grown by molecular beam epitaxy, and a 5.1% laterally expanded Pd (001) lattice was formed. It was shown from investigated magnetic moments that the Fe/Pd interface creates an extra magnetic moment of $0.9\mu_B$/interface atom and that $0.5\mu_B$ of the creation belongs to two adjacent Pd layers and the balance of $0.4\mu_B$ enhances the Fe magnetic moments in the interface, and the magnetic moment of 5Fe/4Pd/10Fe trilayer is equal to the sum of the magnetic moments for the individual 5Fe/8Pd and 8Pd/10Fe bilayers. They concluded that the laterally expanded Pd interlayers are very reluctant to participate in a FM transition even in the presence of the surrounding Fe layers. For thicknesses up to 4 ML the Pd was ferromagnetic throughout the whole layer, and one additional Pd ML destroyed the ferromagnetism of the Pd. However, it possessed a fluctuating magnetic moment partly polarized by the exchange field from adjacent Fe layers.

By using the FMR, BLS, and the surface magneto-optical Kerr effect, Gelinski's group[17,21] also found that the exchange coupling between two Fe films separated by a Pd interlayer whose thicknesses range from 4 to 12 ML is ferromagnetic and exhibits an oscillatory behavior superimposed on the monotonically decreasing background. The periodicity of the oscillatory behavior was approximately 4 ML, and the crossover from FM to a weak AFM exchange coupling occurred at a 12 ML thickness and the AFM exchange coupling showed a maximum value around thicknesses of 14 ML. The observation of a weak AFM exchange coupling in an Fe/Pd/Fe/Ag(001) trilayer is particularly interesting in both the fundamental and application viewpoints.

On the other hand, Childress *et al.*[18] investigated the magnetic properties of high-quality epitaxial Fe/Pd (001) superlattices grown upon MgO (100) substrates. The Pd interfaces were strongly polarized by the proximity of the Fe moments, and the optical Kerr domain imaging observations suggested that the coupling is ferromagnetic for the range of 10–50 Å Pd thicknesses, while the presence of a weak in-plane uniaxial anisotropy in addition to the cubic anisotropy of Fe can be mistaken for appearance of the AFM exchange coupling. Hicken *et al.*[22] measured the exchange coupling at room temperature in epitaxial Fe/Pd/Fe trilayers grown upon MgO(100) substrates by using BLS and the polar magneto-optic Kerr effect, and concluded that the FM exchange coupling is observed for the range of 14–30 Å Pd thickness, although it is of course possible that the AFM exchange coupling may exist for larger Pd interlayer thickness.

The induced spin polarization of individual Pd layer in Co/Pd superlattices was calculated using the layer Koringa-Kohn-Rostoker method by Victora and MacLaren,[23] and also using the discrete variational Slater's $X\ \alpha$ potential by Miura *et al.*[24] In those calculations, the polarization of reverse direction to its neighboring one was surprisingly predicted at the central Pd layer when the Pd layer thickness is 5 ML.

There has already been an attempt to make Pd layer ferromagnetic by expanding its volume in the structure of epitaxial metal film sandwiches. Brodsky and Freeman[25] studied $A(001)/\mathrm{Pd}(001)/A(001)$ sandwiches, where the A represents Ag or Au and the thicknesses of Pd are 7–11 ML. They tried to trigger the FM state in Pd by stretching the Pd lattice by about 2.4% using $A(001)$ templates and found that the samples show an enormous enhancement of the Pauli susceptibility, but the Pd layer still remains paramagnetic. The enormous enhancement of the Pauli susceptibility in the above samples was successfully explained phenomenologically by considering the change of the electronic structure between states of a thin film and a bulk and that of the intra-atomic Coulomb interaction due to the volume expansion and also the electronic Grüneisen constant.[26]

The magnetic properties of superlattices containing Pt interlayers have also been observed. Parkin[4] reported no AFM exchange coupling in Co/Pt superlattices, but alluded to the possibility that structural defects could obscure the presence of the AFM exchange coupling. Dang *et al.*[27] observed conclusive evidence for the AFM exchange coupling in molecular beam epitaxy grown Co/Pt (111) superlattices, consisting of 35- and 52-Å Co and 18-Å Pt layers, by using both nuclear magnetic resonance and low-temperature magnetization measurements. In explanation of the AFM exchange coupling in Co/Pt superlattices, they considered that the RKKY interaction is expected to be more apparent since the e-e exchange interaction among $5d$ conduction electrons in the Pt interlayer is rather weak.

Apart from the induced spin polarization by the proximity of FM layers and the oscillatory exchange coupling between FM layers, some Pd or Pt-based superlattices have another attractive phenomenon, i.e., a perpendicular magnetic anisotropy with a high coercive force. Since the perpendicular magnetic anisotropy at room temperature was reported by Carcia *et al.*[28] for sputtered Pd/Co superlattices with Co thickness less than 8 Å, much attention has been paid to the phenomenon because of interest in the underlying mechanism and applications to high-density magneto-optic recording media. It was found that the systems such as Pd/Co,[16,19,28–31] Pt/Co,[30,32] Au/Co,[33–35] and Ru/Co,[36] containing FM layers thinner than several ML, show perpendicular magnetic anisotropy, but the Ag/Co (Refs. 34, 35, and 37) and Pd/Fe (Refs. 16 and 19) systems do not. Although the origin of the perpendicular magnetic anisotropy is not yet well revealed, it has been generally understood[28] that the appearance of the perpendicular magnetic anisotropy is the result of competition between the Néel-type magnetic surface anisotropy which favors a perpendicular magnetization and a volume anisotropy which favors an in-plane magnetization. Here the volume anisotropy consists of contributions from demagnetization, magnetoelastic, and magnetocrystalline anisotropy energies.

The exchange coupling between FM layers is of great interest if the FM layers are likely to polarize the NM spacer layer magnetically as is the case with Pd and Pt. The purpose in the present article is to calculate the exchange coupling including the effects of a strong e-e exchange interaction among d electrons in the NM spacer layer and to reveal the possibility of the presence of the AFM exchange coupling in such a system.

II. FORMULATION

We derive an expression for the exchange coupling between two FM layers embedded in a NM metal having large exchange enhancement in the paramagnetic susceptibility. We assume that atoms inside FM layers are coupled by the usual exchange interaction as in bulk metal, and only the interface atoms of a FM layer are coupled to the interface atoms of the other FM layer through the spin polarization of conduction electrons in the NM layer. Then, the system is equivalent to two parallel FM planes embedded in a bulk NM metal.[9–12,38,39]

To calculate the exchange coupling between two FM layers, we use the Fermi contact interaction between spins of magnetic atoms and conduction electrons only at interfaces. Then, the perturbation Hamiltonian H' is given by

$$H' = -Jv[\delta(\mathbf{r}-\mathbf{R}_1)\boldsymbol{\sigma}\cdot\mathbf{S}_1 + \delta(\mathbf{r}-\mathbf{R}_2)\boldsymbol{\sigma}\cdot\mathbf{S}_2], \tag{1}$$

where J is a coupling constant, v an atomic volume, $\mathbf{S}_i$ the magnetic spin at site $\mathbf{R}_i$ on interfaces i ($i=1$ and 2), and $\boldsymbol{\sigma}$ the Pauli spin matrices of conduction electrons at position $\mathbf{r}$. Here the $\mathbf{R}_i$'s are designated as $\mathbf{R}_i=(\mathbf{R}_{i\parallel},R_{iz})$.

Before calculating the exchange coupling between magnetic spins $\mathbf{S}_1$ and $\mathbf{S}_2$ embedded in a NM metal having strong e-e exchange interaction, the spin polarization of the conduction electrons surrounding a magnetic spin must be obtained in the NM metal. Giovannini, Peter, and Schrieffer[40] showed numerically that the effect of the e-e exchange interaction among conduction electrons strongly modifies the form of the induced spin polarization surrounding a localized moment in Pd and that the main effect is to increase the polarization on the sites neighboring the localized moment. On the other hand, Moriya[41] obtained an asymptotic expansion for the spin polarization at a long distance from a magnetic spin in the random-phase approximation (RPA) for the e-e exchange interaction.

According to earlier calculations in which an electron gas model was taken,[40,41] the exchange coupling energy $\delta F_{\text{spin pair}}$ between magnetic spins $\mathbf{S}_1$ and $\mathbf{S}_2$ can be calculated within the RPA by

$$\delta F_{\text{spin pair}} = \frac{9}{2}\frac{J^2}{\varepsilon_F}\left(\frac{N_c}{N}\right)^2 G(2k_F|\mathbf{R}_1-\mathbf{R}_2|)\mathbf{S}_1\cdot\mathbf{S}_2, \tag{2}$$

where

$$G(\xi) = \frac{1}{\xi}\frac{\partial}{\partial\xi}\int_{-\infty}^{\infty}\frac{e^{ix\xi}f(x)}{1-(\alpha/2)f(x)}\,dx, \tag{3}$$

with

$$f(x) = 1 + \frac{1-x^2}{2x}\ln\left|\frac{1+x}{1-x}\right|. \tag{4}$$

Here k_F and ε_F are the Fermi wave number and the Fermi energy of conduction electrons, respectively, N_c and N are the numbers of conduction electrons and atoms of the system, respectively, and α is the e-e exchange interaction parameter in the NM spacer layer and its value is defined as $0\leqslant\alpha<1$. The condition of $\alpha\geqslant1$ leads to a spontaneous transition to FM order.

The integral in Eq. (3),

$$F(\xi) \equiv \int_{-\infty}^{\infty}\frac{e^{ix\xi}f(x)}{1-(\alpha/2)f(x)}\,dx, \tag{5}$$

seems too difficult to calculate exactly. However, as explained in detail in the Appendix, by applying the method of contour integrals in the complex plane to $F(\xi)$, we attain the following asymptotic expansion for a long distance between magnetic spins:

$$F(\xi) \approx \frac{2\pi}{(1-\alpha/2)^2}\left[\frac{\sin\xi}{\xi^2} - \frac{\cos\xi}{\xi^3} + \frac{4\alpha}{2-\alpha}\times\left(\frac{3}{2}-\ln 2-\gamma-\ln\,\xi\right)\frac{\cos\xi}{\xi^3}\right] + 2\pi g(\alpha)e^{-y_0\xi}, \tag{6}$$

with

$$g(\alpha) = \left(\frac{2}{\alpha}\right)^2\frac{y_0}{1-(2/\alpha)(1-\alpha/2)[(1-y_0^2)/(1+y_0^2)]} \tag{7}$$

and a solution y_0 of the equation

$$\frac{2}{\alpha}\left(1-\frac{\alpha}{2}\right) = \frac{1+y_0^2}{y_0}\tan^{-1}y_0, \tag{8}$$

where γ=0.5772... is Euler's constant. The second term in Eq. (6) is a contribution from the pole on the imaginary axis, iy_0, in the complex plane, and it has an exponentially decreasing behavior with respect to ξ.

For the special cases of $\alpha\ll1$ or $\alpha\lessapprox1$, y_0 and $g(\alpha)$ can be approximately given as

$$y_0 = \frac{4}{\alpha\pi}, \quad g(\alpha) = \frac{32}{\alpha^3\pi^2}\frac{1}{y_0}, \quad \alpha\ll1, \tag{9}$$

or

$$y_0 = \sqrt{3\left(\frac{1}{\alpha}-1\right)}, \quad g(\alpha) = \frac{3}{\alpha^2}\frac{1}{y_0}, \quad \alpha\lessapprox1. \tag{10}$$

From the $G(\xi)$ of Eq. (3) together with the $F(\xi)$ of Eq. (6), we can obtain the exchange coupling energy between magnetic spins in a NM metal having strong e-e exchange interaction. It is easily seen that the spin polarization distribution surrounding a magnetic spin obtained earlier by Moriya[41] leads to the same result with the $\delta F_{\text{spin pair}}$ of Eq. (2).

So far, we have calculated only the exchange coupling between magnetic spins situated on an arbitrary site at each interface between the FM and NM layers. In order to evaluate the exchange coupling between two FM layers separated by the NM spacer layer, we start from Eq. (2) and sum over all magnetic spin pairs between two FM layers. The summation can be done analytically in the continuous limit of the magnetic spin distribution.

Since $|\mathbf{R}_1-\mathbf{R}_2|$, the distance between magnetic spins $\mathbf{S}_1$ and $\mathbf{S}_2$, is written as $|\mathbf{R}_1-\mathbf{R}_2|=\sqrt{R_\perp^2+(R_{1\parallel}-\mathbf{R}_{2\parallel})^2}$ with the thickness of the NM spacer layer $R_\perp$, one obtains

$$\int\int d\mathbf{R}_{1\parallel}d\mathbf{R}_{2\parallel}G(2k_F\sqrt{R_\perp^2+(\mathbf{R}_{1\parallel}-\mathbf{R}_{2\parallel})^2})$$
$$=\int\int d\mathbf{R}_{1\parallel}d\mathbf{R}_{2\parallel}\int_0^\infty du G(2k_F\sqrt{R_\perp^2+u})\times\delta(u-(\mathbf{R}_{1\parallel}-\mathbf{R}_{2\parallel})^2)$$
$$=\int_0^\infty du G(2k_F\sqrt{R_\perp^2+u})\pi A, \tag{11}$$

where δ means the Dirac delta function and A is the area of the FM layer. Here we used a simple relation

$$\int\int d\mathbf{R}_{1\parallel}d\mathbf{R}_{2\parallel}\delta(u-(\mathbf{R}_{1\parallel}-\mathbf{R}_{2\parallel})^2)=\pi A,$$

which is obtained easily by changing the integral variables.

Let us proceed with the integral by introducing a new variable ξ as $\xi=2k_F\sqrt{R_\perp^2+u}$. By changing the integral variable from u to ξ, Eq. (11) is calculated as

$$\int_0^\infty du\ G(2k_F\sqrt{R_\perp^2+u})\pi A=\int_{2k_FR_\perp}^\infty \frac{1}{2k_F^2}\ \xi\ d\xi\ G(\xi)\pi A$$
$$=\int_{2k_FR_\perp}^\infty \frac{1}{2k_F^2}\ \xi\ d\xi\ \frac{1}{\xi}\frac{\partial F(\xi)}{\partial\xi}\ \pi A$$
$$=-\frac{1}{2k_F^2}F(2k_FR_\perp)\pi A, \tag{12}$$

where $F(\infty)=0$ should be noted.

Therefore, the exchange coupling energy per unit area between two FM layers, $\delta F_{\mathrm{interlayer}}$, is given within the RPA by

$$\delta F_{\mathrm{interlayer}}=-\frac{9\pi}{4}\frac{J^2}{\varepsilon_F^2}\left(\frac{N_c}{N}\right)^2\frac{\hbar^2}{2m}F(2k_FR_\perp)\overline{\mathbf{S}_1}\cdot\overline{\mathbf{S}_2}, \tag{13}$$

where the function $F(\xi)$ has already been given by Eq. (6), and $\overline{\mathbf{S}_1}$ and $\overline{\mathbf{S}_2}$ are the magnetic spin densities on each FM layer, respectively. The formula of Eq. (13) is the main result in the present calculations.

When $\alpha=0$, i.e., no e-e exchange interaction, $\delta F_{\mathrm{interlayer}}$ has already been calculated exactly in another method.[9,39,42] Let us define the exact result by $J_{\mathrm{interlayer}}$. The $J_{\mathrm{interlayer}}$ is given by

$$J_{\mathrm{interlayer}}=-\frac{9\pi}{4}\frac{J^2}{\varepsilon_F^2}\left(\frac{N_c}{N}\right)^2\frac{\hbar^2}{2m}E_x(2k_FR_\perp)\overline{\mathbf{S}_1}\cdot\overline{\mathbf{S}_2}, \tag{14}$$

where

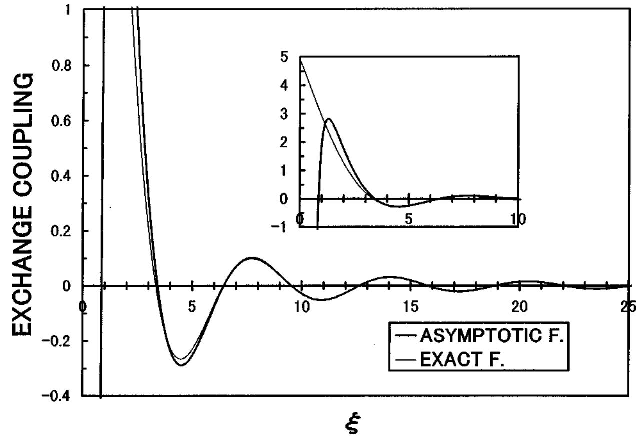

FIG. 1. Exchange coupling (arbitrary units) calculated by the asymptotic $F(\xi)$ of Eq. (6) with $\alpha=0$ and the exact $E_x(\xi)$ of Eq. (15). The positive and negative values correspond to the FM and AFM exchange coupling, respectively. Numerical results by the asymptotic and the exact formulas are shown by solid and light curves, respectively. The vertical scale in an inset is changed in order to see a whole view of results for ξ from 0 to 10. When $\xi=0$, $E_x(\xi)=\pi^2/2\approx4.93$ and $F(\xi)=-\infty$.

$$E_x(\xi)=\pi\left(\frac{\sin\xi-\xi\cos\xi}{\xi^2}+\int_\xi^\infty\frac{\sin t}{t}\,dt\right). \tag{15}$$

When ξ is large enough, we obtain an asymptotic expansion of $E_x(\xi)$ as

$$E_x(\xi)\approx2\pi\left(\frac{\sin\xi}{\xi^2}-\frac{\cos\xi}{\xi^3}\right). \tag{16}$$

When $\alpha\approx0$, on the other hand, the $F(\xi)$ of Eq. (6) becomes equal to the asymptotic formula $E_x(\xi)$ given by Eq. (16) because of the relation $g(\alpha)e^{-y_0\xi}\approx0$, which is easily understood from Eq. (10). Therefore, it is shown that $\delta F_{\mathrm{interlayer}}$ of Eq. (13) and the exactly calculated $J_{\mathrm{interlayer}}$ of Eq. (14) are equal each other in the case of $\alpha\approx0$ and ξ large enough.

III. CALCULATED AND EXPERIMENTAL RESULTS

A. Calculated results

Although the exact $F(\xi)$ in general cases of α could not be obtained because of difficulty of calculations, the asymptotic expansion $F(\xi)$ of Eq. (6) was achieved. Therefore, it is very important in the present theory to find the range of ξ for which the asymptotic expansion $F(\xi)$ of Eq. (6) is valid. Since in the special case of $\alpha=0$ the asymptotic expansion $F(\xi)$ of Eq. (6) can be compared with the exact $F(\xi)$, i.e., $E_x(\xi)$ of Eq. (15), we can determine the suitable range of ξ. The calculated results of the $F(\xi)$ of Eq. (6) with $\alpha=0$ and the $E_x(\xi)$ of Eq. (15) are shown in Fig. 1. The positive and negative values correspond to the FM and AFM exchange couplings, respectively. It can be recognized from Fig. 1 that the asymptotic expansion $F(\xi)$ with $\alpha=0$ is a fairly good approximation to the exact $E_x(\xi)$ of Eq. (15) for ξ larger than about 3 and that the numerical calculations are valid for ξ from the region of the first AFM exchange coupling. Although the results mentioned above were derived

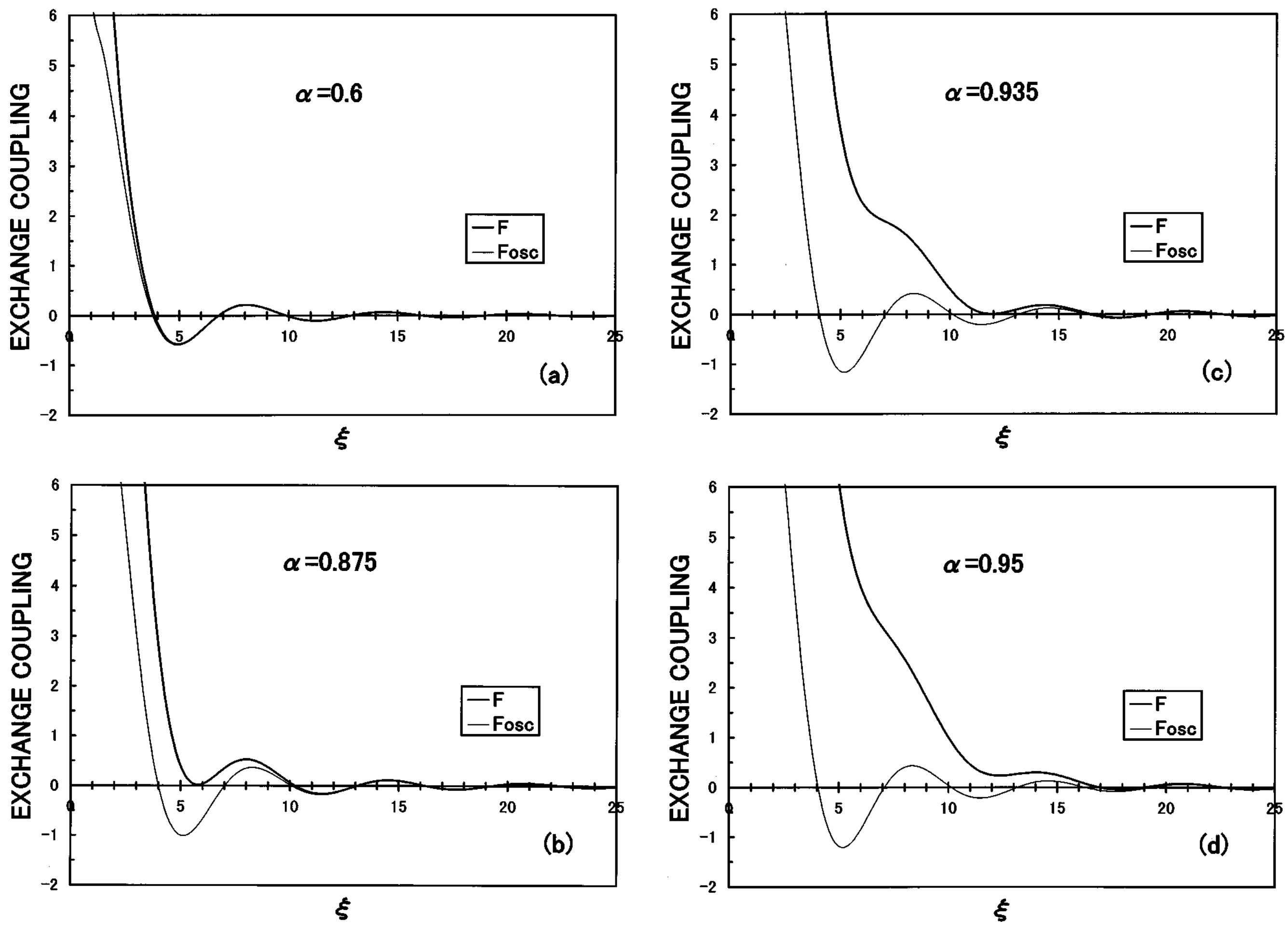

FIG. 2. Calculated results of $F(\xi)$ and $F_{osc}(\xi)$ for various values of α: (a) $\alpha=0.6$, (b) $\alpha=0.875$, (c) $\alpha=0.935$, and (d) $\alpha=0.95$. Solid and light curves indicate results of $F(\xi)$ and $F_{osc}(\xi)$, respectively. The conditions of $\alpha=0.875$ and $\alpha=0.935$ correspond to the borders whether the first AFM exchange coupling appears or not and whether the second AFM exchange coupling appears or not under the disappearance of the first AFM exchange coupling, respectively.

from the examination for the special case of $\alpha=0$, it can be assumed that the $F(\xi)$ with any α of Eq. (6) is sufficiently valid from the region of the first AFM exchange coupling.

Now let us define functions $F_{osc}(\xi)$ and $F_{dec}(\xi)$ by

$$F_{osc}(\xi)\equiv\frac{2\pi}{(1-\alpha/2)^2}\left[\frac{\sin\xi}{\xi^2}-\frac{\cos\xi}{\xi^3}+\frac{4\alpha}{2-\alpha}\times\left(\frac{3}{2}-\ln 2-\gamma-\ln\xi\right)\frac{\cos\xi}{\xi^3}\right]$$

and

$$F_{dec}(\xi)\equiv 2\pi g(\alpha)e^{-y_0\xi}$$

in order to rewrite Eq. (6) as

$$F(\xi)\approx F_{osc}(\xi)+F_{dec}(\xi).$$

Numerical calculations of the $F(\xi)$ were carried out for various values of α, and we show the results for α which give the characteristic behavior in the $F(\xi)$. The calculated results of $F(\xi)$ and $F_{osc}(\xi)$ are shown in Figs. 2(a)–2(d) for $\alpha=0.6$, 0.875, 0.935, and 0.95, respectively. Solid and light curves indicate the results of the $F(\xi)$ and $F_{osc}(\xi)$, respectively. Of course, the difference between the $F(\xi)$ and $F_{osc}(\xi)$, i.e., $F_{dec}(\xi)$, gives the ferromagnetic bias due to effects of the e-e exchange interaction in the NM spacer layer. It is clearly shown that the contributions from the $F_{dec}(\xi)$ to the $F_{osc}(\xi)$ are very important, and the resultant oscillatory behavior, i.e., $F(\xi)$, is drastically changed with increasing α.

B. Comparison between experimental and calculated results

The dependence of the exchange coupling on interlayer thicknesses for magnetic sandwiches Fe/Pd/Fe at 77 K by Gelinski's group[17,21] was compared with calculated results for $\alpha=0.935$, and the results are shown in Fig. 3. Open circles and solid curve indicate experimental and calculated results, respectively. Since the experimental results have shown just disappearance of the region of the second AFM exchange coupling, $\alpha=0.935$ was used in the numerical calculations according to the results shown in Fig. 2(c). The fitting of adjustable parameters in the theory has been made in order to obtain an overall agreement between experimental and calculated results. As a result of the fitting, the calculated results show a well-satisfied agreement with the experimental ones.

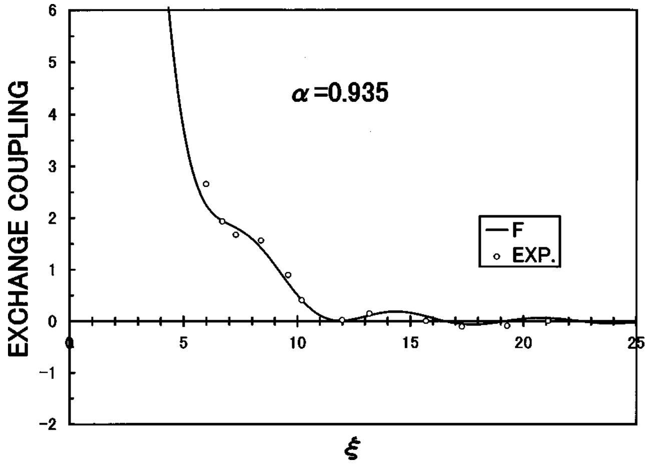

FIG. 3. Experimental and calculated results of exchange coupling (arbitrary units) in magnetic sandwiches Fe/Pd/Fe. Open circles and the solid curve indicate experimental results at 77 K (Refs. 17 and 21) and calculated ones, respectively. $\alpha=0.935$ is used in the numerical calculation.

Moriya used the value of $\alpha=0.95$ in the analysis for the extension of the positive spin polarization in Fe-Pd alloys.[41] Therefore, $\alpha=0.935$ used here seems to be appropriate to the Pd interlayer.

IV. CONCLUSION AND DISCUSSION

By extending the theory of the exchange coupling between two magnetic spins embedded in a NM metal, in which the RPA was used for the e-e exchange interaction, we calculated the exchange coupling between two FM layers mediated through a NM spacer metal having large exchange enhancement in the paramagnetic susceptibility. The formula of the exchange coupling is given by Eq. (13) together with Eq. (6). The present theory revealed that the exchange coupling decreases with an increasing interlayer thickness in a nontrivial manner, and an oscillatory behavior superimposed on the exponentially decreasing background is clearly shown. The exponentially decreasing background produces a ferromagnetic bias to the oscillatory exchange coupling remarkably for low spacer thicknesses.

We can see a series of changes of the oscillatory behavior of $F(\xi)$ with increasing α from Figs. 2(a)–2(d). It is found from Figs. 2(b) and 2(c) that the condition $\alpha=0.875$ corresponds to a border whether the first AFM exchange coupling appears or not, and the condition $\alpha=0.935$ corresponds to a border whether the second AFM exchange coupling appears or not. The first AFM exchange coupling can appear up to $\alpha=0.875$, and the first AFM exchange coupling disappears and the second AFM exchange coupling can still appear for α above 0.875 and below 0.935. Moreover, both the first and second AFM exchange couplings disappear for α above 0.935. However, the traces of the disappeared AFM exchange couplings can be recognized as an easy slope and a dip in the region of the FM oscillatory exchange coupling, e.g., an easy slope at $\xi\approx7$ and a dip at $\xi\approx12$ for $\alpha=0.95$ as shown in Fig. 2(d).

From Fig. 3 we see that the first and second AFM exchange couplings disappear and the third AFM one appears in magnetic sandwiches, Fe/Pd/Fe by Gelinski's group,[17,21] and the present theoretical result agrees well with the experimental results. An easy slope at $\xi\approx7$ in the experimental results suggests a trace of the first AFM exchange coupling.

Moriya[41] has estimated the value of α for Pt metal by comparing the magnetic susceptibility with the electronic specific data and obtained $\alpha=0.6$. It is small compared with that of Pd metal. It is seen from the results shown in Fig. 2(a) that the contribution from the exponentially decreasing behavior is fairly reduced in the case of magnetic superlattices with a Pt interlayer. Therefore, the conventional oscillatory exchange coupling is dominant and the first and second AFM exchange coupling can be definitely expected as Dang *et al.*[27] observed the conclusive evidence for AFM exchange coupling in Co/Pt superlattices.

We considered a highly simplified model of an interacting free electron gas to describe the NM spacer metal. Therefore, it should be noted that the results in the present article provide a partial and qualitative understanding of real systems. In transition metals such as Pd or Pt near the right end of the transition-metal series, the Fermi level lies within the d band, and the Fermi surface consists of two regions of holes and one compensating region of electrons.[43] Of these three surfaces, the small pockets of heavy d-like holes at X are ellipsoids of revolution prolate along the ΓX direction. Indeed, it has been found[21] that the X-centered hole pockets along the ΓX direction in fcc Pd can lead to RKKY-like oscillations in the exchange coupling in Fe/Pd/Fe trilayers.

Since it was difficult to take account of the band structure of Pd and Pt precisely in the theory, we made use of an effective mass approximation for the d-like holes and a model of an interacting free electron gas was applied to residual d-like holes in the present calculations. This model has been used in the earlier calculations for the spin polarization of the conduction electrons surrounding a magnetic spin in Pd metal.[40,41] This picture for conduction electrons in the NM spacer metals simplifies our calculations a great deal while at the same time retaining the essential physics of the problem. Therefore, we could perform the calculations analytically and obtain a physically transparent illustration of the various aspects of the exchange coupling. However, some changes in the present results may be expected in the calculated results due to band structure effects since the oscillation periods, amplitudes, and phases are dependent on the band structure near the Fermi surface of the spacer transition metal.

As we have seen in the preceding sections, the present model for the spacer transition metals, though being extremely simplified one, seems to explain essential features of the exchange coupling.

It was revealed that if the e-e exchange interaction among d electrons in the NM spacer layer is strong, it plays a crucial role in the interlayer thickness dependence of the exchange coupling between FM layers in superlattices.

APPENDIX

In this appendix we explain the method of integral of $F(\xi)$ of Eq. (5). Let us consider a contour integral of the type

$$\oint \frac{e^{iz\xi}f(z)}{1-(\alpha/2)f(z)}\,dz, \tag{A1}$$

where $f(z)$ is identical to Eq. (4). The integral is performed in the z complex plane by closing the path, consisting of lines $(-\infty, -1)$, $(-1, 1)$, and $(1, \infty)$ on the real axis, and infinitesimal semicircles centered at $z=\pm 1$ and infinite one centered at $z=0$ in the upper half-plane. The integral along the semicircle centered at $z=0$ and the integrals along the semicircles centered at $z=\pm 1$ in the upper half-plane vanish as the radius tends to infinity and infinitesimal, respectively. The phase of $\ln(1+z)/(1-z)$ is π for the domains $(-\infty, -1)$ and $(1, \infty)$ and 0 for $(-1, 1)$. A pole of the first order occurs on the imaginary axis at $z=iy_0$. Therefore, the contour integral gives

$$2\pi i\times(\text{residue at } iy_0)=\left\{\int_{-\infty}^{-1}+\int_{1}^{\infty}\right\}dx\left\{\frac{e^{ix\xi}\left(f(x)+i\pi\frac{1-x^2}{2x}\right)}{1-\frac{\alpha}{2}\left(f(x)+i\pi\frac{1-x^2}{2x}\right)}\right\}+\int_{-1}^{1}\frac{e^{ix\xi}f(x)}{1-\frac{\alpha}{2}f(x)}dx$$

$$=\left\{\int_{-\infty}^{-1}+\int_{1}^{\infty}\right\}dx\left\{\frac{e^{ix\xi}\left(f(x)+i\pi\frac{1-x^2}{2x}\right)}{1-\frac{\alpha}{2}\left(f(x)+i\pi\frac{1-x^2}{2x}\right)}-\frac{e^{ix\xi}f(x)}{1-\frac{\alpha}{2}f(x)}\right\}+\int_{-\infty}^{\infty}\frac{e^{ix\xi}f(x)}{1-\frac{\alpha}{2}f(x)}dx. \quad \text{(A2)}$$

After some calculations, we obtain

$$\int_{-\infty}^{\infty}\frac{e^{ix\xi}f(x)}{1-\frac{\alpha}{2}f(x)}dx=-\frac{2}{\alpha}\left\{\int_{-\infty}^{-1}+\int_{1}^{\infty}\right\}dx\times\left\{\frac{e^{ix\xi}}{1-\frac{\alpha}{2}\left(f(x)+i\pi\frac{1-x^2}{2x}\right)}-\frac{e^{ix\xi}}{1-\frac{\alpha}{2}f(x)}\right\}+2\pi i\times(\text{residue at } iy_0). \quad \text{(A3)}$$

The first term in Eq. (A3) seems too difficult to calculate exactly. However, it is possible to calculate the term in an asymptotic expansion for large ξ.[44]

The pole of the first order iy_0 can be obtained by solving the equation $1-(\alpha/2)f(iy_0)=0$, i.e.,

$$\frac{2}{\alpha}\left(1-\frac{\alpha}{2}\right)=\frac{1+y_0^2}{y_0}\tan^{-1}y_0. \quad \text{(A4)}$$

The residue at iy_0, res (iy_0), is given by

$$\text{res}(iy_0)=-i\left(\frac{2}{\alpha}\right)^2\frac{y_0e^{-y_0\xi}}{1-(1-y_0^2)\frac{\tan^{-1}y_0}{y_0}}=-i\left(\frac{2}{\alpha}\right)^2\frac{y_0e^{-y_0\xi}}{1-\frac{2}{\alpha}\left(1-\frac{\alpha}{2}\right)\frac{1-y_0^2}{1+y_0^2}}, \quad \text{(A5)}$$

where Eq. (A4) is used for deriving the equation in the second line.

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