Noncollinear interlayer coupling across a semiconductor spacer

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Based on the extended s-d exchange model, which includes both isotropic and anisotropic spin interactions between conduction electrons and local states, we have derived analytically the interlayer coupling across a semiconductor spacer with a general band structure. Both Heisenberg-type and Dzyaloshinski-Moriya (DM) –type Ruderman-Kittel-Kasuya-Yosida–like interlayer coupling are obtained as a result of spin-orbit interaction. The interlayer coupling decreases exponentially with spacer thickness and the oscillation period depends on the band structure and orientation of spacers. Our result is different from previous theory; in particular, the DM-type interlayer exchange coupling offers a natural explanation to the noncollinear alignment of neighboring ferromagnetic layers as were observed in recent experiments on magnetic-semiconductor multilayer structures. [S0163-1829(97)00447-5]

Indirect exchange interaction between ferromagnetic layers across semiconductor spacers has been extensively studied both experimentally and theoretically. In experiments, the following different features were observed in different structures: ferromagnetic interlayer coupling for Fe/SiO/Fe (Ref. 1) and Fe/Ge/Fe (Ref. 2) sandwiches and spacer-layer-dependent oscillatory behavior of interlayer coupling for Fe/FeSi (Ref. 3) and Fe/Si (Ref. 4) trilayers and multilayers. For some structures the coupling is antiferromagnetic at room temperature and ferromagnetic or no coupling at low temperature.[3] In a recent experiment[5] the interlayer coupling in Fe/SiFe/Fe was found to be strongly antiferromagnetic and decrease exponentially with spacer thickness. The noncollinear interlayer coupling is also observed in Fe/FeSi (Ref. 6) multilayers.

Various theoretical models were proposed to explain the origin of the interlayer exchange coupling across semiconductor spacers. The first is a tunneling model[7,8] for interlayer coupling. Here, the spacer layer was taken as a potential barrier and the problem was solved within the picture of free electrons. The interlayer coupling was predicted to be an exponentially decaying function of spacer thickness z and takes the form $J=z^{-2}\exp(-z/\lambda_d)$, where λ_d is a characteristic length determined by the potential barrier. The effect of indirect tunneling on the interlayer coupling through the localized states was also studied recently.[9] The second is the Kondo insulator model.[10] The interlayer coupling calculated in this model oscillates from ferromagnetic to antiferromagnetic as the spacer thickness varies. The exponential decaying behavior was also obtained, but with a constant prefactor $J=\exp(-z/\lambda_d)$. The third model[11] is based on the indirect magnetic exchange interaction mediated by bound exciton and correlated intermediate states. The attractive Coulomb interaction between an electron in the conduction band and a hole in the valance band of a spacer was believed to play an important role.

All the present approaches did not consider the Ruderman-Kittel-Kasuya-Yosida (RKKY) –like indirect exchange interaction since it is believed that the result is not relevant to the temperature dependence of interlayer coupling observed in the experiments. However, a recent study[12] on the magnetic-semiconductor multilayer structure reveals a different type of temperature dependence of interlayer coupling. The measured saturation field suggests that interlayer coupling becomes weaker as temperature increases, which is in disagreement with the theoretical predictions of current models and thus a different approach is needed. The problem of RKKY-like exchange coupling between two magnetic impurities embedded in a semiconductor host was studied long ago by Abrikosov[13] and was applied to the one-dimensional semiconductor-based spin glasses. The calculated physical properties agree qualitatively with the experiment data and they show that the RKKY-like exchange coupling does exist in the system with a semiconductor host.

In this paper we derive analytical results of both Heisenberg-type and Dzyaloshinski-Moriya (DM) –type RKKY-like interlayer coupling across semiconductor spacers with a general band structure. Our study is based on the extended s-d exchange model derived in a previous work,[14] which includes the effect of spin-orbit interaction. The extended s-d exchange model contains both isotropic and anisotropic terms. The anisotropic scattering potential also leads to noncollinear interlayer coupling across semiconductor spacers. The interlayer exchange coupling decreases exponentially as spacer thickness increases and can be expressed as $J=z^{-3/2}\exp(-z/\lambda_d)$, with z and λ_d denoting the spacer thickness and characteristic length determined by the band structure of the semiconductor spacer. Our calculated interlayer coupling can be either monotonic or oscillatory, depending on the band structure and orientation of the spacer. This result is different from those obtained in the previous theories.[8,10]

The Hamiltonian we start with is a sum of the Hamiltonians contributed by the ferromagnetic and semiconductor layers. In the ferromagnetic layer, we adopt the extended Anderson s-d mixing Hamiltonian,[14] which takes into account both the extended s electrons and localized d electrons. The d electrons form the localized magnetic moments in the ferromagnetic layer and the s electrons extend into the semiconductor layer, mediating the magnetic coupling between the neighboring ferromagnetic layers. The extended

s-d mixing Hamiltonian in the ferromagnetic layer can be transformed into an extended s-d exchange Hamiltonian using the Schrieffer-Wolff transformation. The interaction between s electrons and d electrons to the first order of spin-orbit coupling reads[14]

$$V=-\sum_{\vec{k},\vec{k}',n}\frac{1}{2}\psi^{\dagger}_{\vec{k}'}\vec{\sigma}\psi_{\vec{k}}\cdot(J_{\vec{k}'\vec{k}}\vec{S}_n-i\vec{T}_{\vec{k}'\vec{k}}\times\vec{S}_n)e^{i(\vec{k}'-\vec{k})\cdot\vec{R}_n}. \tag{1}$$

Here $\psi_{\vec{k},(dn)}=(C_{\vec{k},(dn)\uparrow},C_{\vec{k},(dn)\downarrow})^T$ and $\vec{S}_n=(1/2)\psi^{\dagger}_{dn}\vec{\sigma}\psi_{dn}$. $C^{\dagger}_{\vec{k}s}$ and $C^{\dagger}_{dns}$ are the creation operators for the s electron with momentum $\vec{k}$ and localized d electron on lattice site n, respectively. s is the spin index. From the expression in Ref. 14, we have $J_{\vec{k}'\vec{k}}=J_{\vec{k}\vec{k}'}$ and $\vec{T}_{\vec{k}'\vec{k}}=-\vec{T}_{\vec{k}\vec{k}'}$. In the high-symmetry bulk crystals, the space inversion symmetry requires $\vec{T}_{\vec{k}'\vec{k}}=\vec{T}_{\vec{k}\vec{k}'}$ and $\vec{T}_{\vec{k}'\vec{k}}=0$. This is not the case in the ferromagnetic-semiconductor multilayer structures where the space inversion symmetry is generally broken, especially near the interface. Thus $\vec{T}_{\vec{k}'\vec{k}}$ can have a nonzero value. As we will see below, this term contributes to the noncollinear interlayer exchange coupling between the neighboring ferromagnetic layers.

To get a physically transparent result, we assume that the localized moment is the same and the summation over n can be done for each ferromagnetic layer. The summation over a single atomic layer in the ferromagnetic layer gives rise to a conservation law for the parallel electronic momentum $\vec{k}'_{\parallel}=\vec{k}_{\parallel}$ and summation over different atomic layers results in an additional factor $g_{\vec{k}',\vec{k}}=\sin[(k'_z-k_z)Na/2]/\sin[(k'_z-k_z)a/2]$. Here N and a are the number of atomic layers in the ferromagnetic layer and the lattice constant, respectively. To simplify the calculation further, we assume that the interface between the ferromagnetic layer and semiconductor layer is transparent. Then the exchange coupling between two ferromagnetic layers across a semiconductor spacer can be obtained using the second-order perturbation theory in terms of V in the same way as in Ref. 8. The effective coupling Hamiltonian can be expressed by

$$H_{eff}=\sum_{n,m}\sum_{\vec{k}_{\parallel}}\sum_{k_z,k'_z}\sum_{s,s'}\frac{\langle\vec{k}s|V|\vec{k}'s'\rangle\langle\vec{k}'s'|V|\vec{k}s\rangle}{\epsilon_{\vec{k}}-\epsilon_{\vec{k}'}}, \tag{2}$$

where n denotes one ferromagnetic layer and m denotes the neighboring layer. $(k_{\parallel},k_z,k'_z)$ is a sum over the conduction and valence bands of the semiconductor. The interlayer coupling energy per area is given by

$$H_{eff}=\frac{1}{2}\sum_{n,m}\sum_{\vec{k}_{\parallel}}\sum_{k_z,k'_z}\frac{f_{\vec{k}}(1-f_{\vec{k}'})e^{i(\vec{k}_z-\vec{k}'_z)z}}{\epsilon_{\vec{k}}-\epsilon_{\vec{k}'}}\times[|J'_{\vec{k},\vec{k}'}|^2\vec{M}_m\cdot\vec{M}_n+2iJ'_{\vec{k},\vec{k}'}\vec{T}'_{\vec{k},\vec{k}'}\cdot(\vec{M}_n\times\vec{M}_m)+(\vec{T}'_{\vec{k},\vec{k}'}\times\vec{M}_m)\cdot(\vec{T}'_{\vec{k},\vec{k}'}\times\vec{M}_n)]. \tag{3}$$

Here z is the spacer layer thickness, $J'_{\vec{k},\vec{k}'}=g_{\vec{k},\vec{k}'}J_{\vec{k},\vec{k}'}$, and $\vec{T}'_{\vec{k},\vec{k}'}=g_{\vec{k},\vec{k}'}\vec{T}_{\vec{k},\vec{k}'}$. $\vec{M}_{n\,(m)}$ is the magnetization of layer $n(m)$ per area. $\vec{k}\equiv(k_{\parallel},k_z)$ and $f_{\vec{k}}$ is the Fermi distribution function. Further analysis requires the band structure of the semiconductor and full knowledge of the matrix elements $J'_{\vec{k},\vec{k}'}$, $\vec{T}'_{\vec{k},\vec{k}'}$. To simplify the calculation, we consider the following band structure:[13] one maximum in the valence band at $\vec{k}=\vec{0}$ and several equivalent minima in the conduction band at $\vec{k}=\vec{K}_i$. The electrons and holes are assumed to be isotropic and the energy spectrum can be expressed as $\epsilon_k=-\hbar^2k^2/2m_h$ for the valence band and $\epsilon_k=\hbar^2(k-K_i)^2/2m_e+E_g$ for the conduction band. The matrix elements are assumed to be slowly varying function of wave vector $\vec{k}$, so that $J'_{\vec{k},\vec{k}'}\equiv J$ and $\vec{T}'_{\vec{k},\vec{k}'}\equiv\vec{\tau}(k_z-k'_z)$, with J and $\vec{\tau}$ a constant and a constant vector, respectively. In our case, $\vec{\tau}$ is taken to be parallel or antiparallel to the z axis.

The expression in square brackets in Eq. (3) can be rearranged as $(J'_{\vec{k},\vec{k}'}\vec{M}_m-i\vec{T}'_{\vec{k},\vec{k}'}\times\vec{M}_m)\cdot(J'_{\vec{k},\vec{k}'}\vec{M}_n+i\vec{T}'_{\vec{k},\vec{k}'}\times\vec{M}_n)$. If $J'_{\vec{k},\vec{k}'}$ and $\vec{T}'_{\vec{k},\vec{k}'}$ were both independent of $\vec{k}$ and $\vec{k}-\vec{k}'$, H_{eff} would be isomorphic to the symmetry of isotropic case, as pointed out by Shekhtman *et al.*[15] However, this is not the case and the degeneracy problem encountered in Moriya's exchange coupling is generally absent in the multilayer system. This point can be seen even more clearly after the momentum integration. The Hamiltonian does not have the feature needed to be transformed into the isotropic case. Thus the exchange interaction through semiconductor spacers is anisotropic in general. Since $|\vec{T}_{\vec{k},\vec{k}'}|/|J_{\vec{k},\vec{k}'}|$ is a small parameter, only the zeroth- and first-order terms are kept below.

As usual, the summation over momentum in Eq. (3) can be replaced by an integration. After the variable transformation $k_z-k'_z=q-K^z_i$ and $(m_e/m_h)^{1/2}k_z+(m_h/m_e)^{1/2}(k'_z-K^z_i)=K$, one obtains at zero temperature

$$H_{eff}=-\frac{1}{4(2\pi)^4}\sum_{m,n,i}\int dk^2_{\parallel}dq\,dK\frac{e^{iqz}e^{-iK^z_iz}}{\sqrt{\frac{m_h}{m_e}}+\sqrt{\frac{m_e}{m_h}}}\times\frac{[J^2\vec{M}_n\cdot\vec{M}_m+2iJ(q-K^z_i)\vec{\tau}\cdot(\vec{M}_n\times\vec{M}_m)]}{\frac{\hbar^2}{2}\frac{K^2+q^2}{m_h+m_e}+\Delta(k_{\parallel})}+\text{c.c.} \tag{4}$$

Here $\vec{K}_i=(K^{\parallel}_i,K^z_i)$ and $\Delta(k_{\parallel})=\hbar^2k^2_{\parallel}/2m_h+\hbar^2(k_{\parallel}-K^{\parallel}_i)^2/2m_e+E_g$.

The integration over q and K is reduced to a Hankel function. After using its asymptotic form $\kappa_\nu(x)=\sqrt{2\pi/x}e^{-x}$, H_{eff} can be rewritten as $H_{eff}=H_1+H_2+H_3$, with

$$H_1=-\frac{\sqrt{m_hm_e}}{2\hbar^2}\sum_{m,n,i}\frac{J^2e^{-iK^z_iz}}{8\pi^3}\int dk^2_{\parallel}\sqrt{\frac{\pi}{2\alpha}}\frac{e^{-\alpha z}}{\sqrt{z}}\vec{M}_n\cdot\vec{M}_m+\text{c.c.}, \tag{5a}$$

$$H_2=\frac{\sqrt{m_hm_e}}{2\hbar^2}\sum_{m,n,i}\frac{Je^{-iK^z_iz}}{8\pi^3}\int dk^2_{\parallel}\sqrt{\frac{\pi\alpha}{2}}\frac{e^{-\alpha z}}{\sqrt{z}}2\vec{\tau}\cdot(\vec{M}_n\times\vec{M}_m)+\text{c.c.}, \tag{5b}$$

$$H_3=\frac{\sqrt{m_h m_e}}{2\hbar^2}\sum_{m,n,i}\frac{Je^{-iK_i^z z}}{8\pi^3}\int dk_\parallel^2\sqrt{\frac{\pi}{2\alpha}}\frac{e^{-\alpha z}}{\sqrt{z}}2iK_i^z\vec{\tau}\cdot(\vec{M}_n\times\vec{M}_m)+\text{c.c.},\tag{5c}$$

where

$$\alpha=\frac{m_h+m_e}{\sqrt{m_h m_e}}\sqrt{\left(k_\parallel-\frac{m_h}{m_h+m_e}K_i^\parallel\right)^2+K_g^2}$$

and

$$K_g^2=\frac{m_h m_e}{(m_h+m_e)^2}K_i^{\parallel 2}+\frac{2}{\hbar^2}E_g\frac{m_h m_e}{m_h+m_e}.$$

The integration over $k_\parallel$ can be done by shifting the origin of $k_\parallel$ to $k_\parallel-[m_h/(m_h+m_e)]K_i^\parallel=0$ and an incomplete Γ function is obtained. Using the asymptotic expression $\Gamma(\alpha,x)=x^{\alpha-1}e^{-x}$, one finally arrives at

$$H_1=-\sqrt{\frac{\pi}{2}}\frac{J^2}{\hbar^2}\frac{(m_h m_e)^{5/4}}{(m_h+m_e)^{3/2}}\sum_{m,n,i}\frac{\sqrt{K_g}}{z^{3/2}}\cos K_i^z z\times\exp\left(-\frac{m_h+m_e}{\sqrt{m_h m_e}}K_g z\right)\vec{M}_n\cdot\vec{M}_m,\tag{6a}$$

$$H_2=\sqrt{\frac{\pi}{2}}\frac{2J}{\hbar^2}\frac{(m_h m_e)^{3/4}}{(m_h+m_e)^{1/2}}\sum_{m,n,i}\frac{\sqrt{K_g}}{z^{3/2}}\cos K_i^z z\times\exp\left(-\frac{m_h+m_e}{\sqrt{m_h m_e}}K_g z\right)K_g\vec{\tau}\cdot(\vec{M}_n\times\vec{M}_m),\tag{6b}$$

$$H_3=\sqrt{\frac{\pi}{2}}\frac{2J}{\hbar^2}\frac{(m_h m_e)^{5/4}}{(m_h+m_e)^{3/2}}\sum_{m,n,i}\frac{\sqrt{K_g}}{z^{3/2}}\sin K_i^z z\times\exp\left(-\frac{m_h+m_e}{\sqrt{m_h m_e}}K_g z\right)K_i^z\vec{\tau}\cdot(\vec{M}_n\times\vec{M}_m).\tag{6c}$$

These expressions are the effective exchange coupling across semiconductor spacers at zero temperature. H_1 is the usual Heisenberg-type RKKY interlayer coupling, while H_2 and H_3 are the DM-type interlayer couplings.

The Heisenberg-type RKKY interlayer exchange coupling across semiconductor spacers reminds one of the coupling across nonmagnetic metallic spacers. While for the metallic spacer the thickness dependence of interlayer coupling is described by $J\propto z^{-2}$, the interlayer coupling across a semiconductor spacer has a much stronger dependence on the spacer thickness and has the form $J\propto z^{-3/2}e^{-z/\lambda_R}$, where $\lambda_R=\sqrt{m_h m_e}/(m_h+m_e)K_g$ is a characteristic length determined by the band structure of spacers. Note that our result also differs from the optical electrons theory of Bruno,[8] where the interlayer coupling takes the form $J\propto z^{-2}e^{-z/\lambda_B}$. This difference is due to the neglect of the $\vec{k}$ dependence of the semiconductor gap and the barrier height having been taken as a constant.[8]

In our model the oscillation period depends on the band structure and orientation of spacers. For a direct-gap semiconductor spacer ($K_i^z=0$), the coupling is always ferromagnetic and no oscillation behavior can be observed. For an indirect-gap semiconductor spacer, the interlayer coupling oscillates from ferromagnetic to antiferromagnetic as spacer thickness increases. The oscillation period is given by $2\pi/K_i^z$ and depends on the orientation of spacers. Interlayer coupling does not oscillate for the special orientation with $K_i^z=0$. For a typical indirect-gap Si semiconductor spacer with lattice constant $a=5.4$ Å, $K_i^z=\pi/a$ for (001) orientation and the oscillation period is $d=2a=10.8$ Å. This oscillation period is much larger than the interface roughness. Indeed, a clear oscillation period of 10 Å has been observed in Fe/Si multilayered structures.[5]

As in the case of metallic spacer, the spin-orbit interaction also induces DM-type interlayer coupling in the ferromagnetic-semiconductor multilayer structures. To estimate the relative strength of DM coupling over Heisenberg coupling, we use the band structure parameters of Si and $H_2/H_1\approx(2\tau/J)[2.2\sqrt{2mE_g/\hbar^2}]$. τ depends on the degree of symmetry breaking near the ferromagnetic-semiconductor interface and its order of magnitude can be only roughly estimated. In the Fe/Cu multilayers, it is more or less known[14] that $(2\tau/J)[\sqrt{2mE_F/\hbar^2}]\approx 0.1$. If one assumes that τ is roughly the same, we obtain $H_2/H_1\approx 0.09$ and H_3/H_1 has a similar order of magnitude. For direct-gap semiconductor spacers $H_3=0$ and the DM-type H_2 and the Heisenberg-type H_1 interlayer couplings depend on the spacer thickness in exactly the same way. The larger the energy gap, the larger the ratio H_2/H_1. For an indirect energy gap $H_3\neq 0$. This term has the same function dependence on spacer thickness as that of H_1, except for a phase difference of $\pi/2$. H_3 reaches its maximum when H_1 and H_2 are near zero. It is interesting to compare the oscillatory behavior of DM interlayer coupling across semiconductor spacers with that across metallic spacers. In zero temperature, DM interlayer coupling across metallic multilayers has only one term; its phase differs from Heisenberg interlayer coupling by $\pi/2$. In the structure with semiconductor spacers, DM interlayer coupling has two terms. One term is the same as that in the metallic layered structure, whereas another term has the same spacer thickness dependence as that of a Heisenberg term. So in the magnetic layered structure with semiconductor spacers, the DM term does not reach its largest value as the Heisenberg term vanishes.

The spacer thickness fluctuation theory[16] of biquadratic coupling in metallic multilayers cannot be applied to the multilayers with semiconductor spacers. This is so since in metallic multilayers the RKKY interaction oscillation period is about 2 or 3 Å. The long-period oscillation of interlayer coupling is due to the alias effect, so one or two atomic-size changes in the distance between magnetic impurities induces a sign change of the RKKY coupling. Also, the fluctuation in the spacer thickness results in competition between ferromagnetic and antiferromagnetic interlayer coupling, thus the magnetization of ferromagnetic layers will align noncollinearly. However, in the multilayers with semiconductor spacers, the oscillation period of coupling between magnetic impurities is about 10 Å, so the sign does not change on an atomic scale. The fluctuation of the spacers thickness does not induce the biquadratic coupling in the multilayers with semiconductor spacers.

In summary, we have derived analytical results of both Heisenberg-type and DM-type RKKY-like interlayer coupling across semiconductor spacers with a general band structure. The interlayer exchange coupling decreases exponentially as spacer layer increases and is expressed as $J = z^{-3/2}\exp(-z/\lambda_d)$. For a direct-gap semiconductor spacer, the coupling is always ferromagnetic and no oscillation behavior can be observed. For an indirect-gap semiconductor spacer, the interlayer coupling oscillates from ferromagnetic to antiferromagnetic as spacer thickness increases. The oscillation period is also influenced by the orientation of spacers. Furthermore, the noncollinear alignment of neighboring ferromagnetic layers can be naturally explained by our DM interlayer coupling.

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