Impurity-Assisted Interlayer Exchange Coupling across a Tunnel Barrier

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Localized impurity or defect states in the insulating barrier layer separating two ferromagnetic films affect dramatically the interlayer exchange coupling (IEC), making it significantly stronger compared to perfect barriers. We demonstrate that the impurity-assisted IEC becomes antiferromagnetic if the energy of the impurity states matches the Fermi energy and that the coupling strength decreases with temperature. These results explain available experimental data on the IEC across tunnel barriers.

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Magnetizations of two ferromagnetic (FM) films separated by a thin insulating barrier layer are exchange coupled due to the tunneling spin polarization propagating across the barrier [1]. The interlayer exchange coupling (IEC) energy per unit area is given by $E_{\mathrm{IEC}} = -J\cos\theta$, where θ is the angle between the magnetizations of the two ferromagnetic layers. Positive values of the coupling constant J favor parallel alignment of the magnetizations, and negative values favor antiparallel alignment. The magnitude and sign of the IEC depends on the barrier height, the Stoner exchange splitting of the ferromagnets, and temperature [1,2]. Unlike a metallic spacer layer, the tunneling barrier leads to nonoscillatory coupling which decays exponentially as a function of the barrier thickness reflecting the evanescent nature of the exchange-mediated states (for a recent review on IEC, see Ref. [3]).

Experimental observation of the IEC across a tunnel barrier is much more demanding compared to metallic spacers. For amorphous insulators like Al_2O_3, which are widely used in tunneling magnetoresistance (TMR) studies [4], the IEC has not been observed experimentally. Recently, however, Faure-Vincent *et al.* found a strong antiferromagnetic (AF) coupling in epitaxial Fe/MgO/Fe(001) structures [5]. For a thin MgO layer of 6 Å they measured $J \approx -0.26\ \mathrm{erg/cm^2}$, which is of the same order of magnitude as the IEC across metallic spacers [3]. The coupling strength decreased exponentially with the barrier thickness reflecting the tunneling mechanism of the coupling. Even a stronger AF IEC, up to 2 $\mathrm{ergs/cm^2}$, was found by Gareev *et al.* [6] and Bürgler *et al.* [7] in epitaxial Fe/Si/Fe(001) trilayers. Transport measurements indicated that the Si spacer layer is nonconductive and acts as a tunnel barrier of a height of several tenths of an eV, supporting the tunneling mechanism for the IEC. A strong AF coupling exponentially decreasing on a scale less than 2 Å was also found in the epitaxial Fe/Si/Ge/Si/Fe system [8].

The sizable values of the IEC found experimentally are surprising in a view of the expected strong exponential decay of the coupling, especially for MgO representing a high potential barrier [9]. Also the origin of the AF exchange is not evident. According to Slonczewski's model [1], the IEC changes sign and becomes AF for a relatively low potential barrier. This fact was exploited by Faure-Vincent *et al.* [5] who fitted the magnitude and thickness dependence of the AF coupling in epitaxial Fe/MgO/Fe structures. Unfortunately, a more accurate analysis shows that for the parameters used by Faure-Vincent *et al.* the Slonczewski's formula which was derived in the asymptotic limit of a thick barrier is valid only for barrier thicknesses above 12 Å. A proper integration over transverse momenta and energies of incident electrons results in a *ferromagnetic* exchange up to about 10 Å [10].

Bürgler *et al.* [7] found it impossible to fit their experimental data on the IEC in epitaxial Fe/Si/Fe structures using a physically reasonable set of parameters within the Slonczewski's formula. Also their results on temperature dependence of the coupling are in disagreement with available models. In particular, the quantum interference model [2] predicts an increase in the IEC with temperature due to the thermal population of the excited electronic states which experience a lower tunneling barrier. It is, however, found experimentally that the coupling strength decreases with temperature [7].

In this Letter we show that all these inconsistencies can be explained within a model which assumes that the barrier is not perfect but contains impurities or defects creating localized states within the band gap of the insulator. The resonant origin of the IEC induced by these localized states leads to a significant enhancement in the coupling. We demonstrate that the impurity-assisted IEC becomes AF if the energy of the impurity states matches the Fermi energy and that the coupling strength decreases with temperature which is consistent with the experimental observations.

We calculate the IEC between two seminfinite FM layers separated by a plane barrier layer of thickness d. The electronic structure of the ferromagnets is modeled by free-electron spin-split bands of the exchange splitting parameter Δ_{ex}. The barrier is represented by a rectangular potential of height U_b. An impurity is modeled by a delta-function potential of depth U_i located in the barrier at distance r_i from the left interface. This potential creates a quantum well containing an impurity level of energy E_i. We assume that the y axis lies perpendicular to the planes. The magnetization of the left ferromagnet is pointed along the z axis, and the magnetization of the right ferromagnet is canted with angle θ with respect to the z axis and lies in the plane on the layer. The single particle Schrödinger equation for this system is written as follows:

$$\left[-\frac{\hbar^2}{2m}\nabla^2 + V_0^{(n)}(\mathbf{r}) + V_{ex}^{(n)}(\mathbf{r}) + V_i(\mathbf{r})\right]\psi(\mathbf{r}) = E\psi(\mathbf{r}). \quad (1)$$

Here n is the layer index: $n = 1$ and $n = 3$ denote the left and right ferromagnets, and $n = 2$ denotes the barrier. $\psi(\mathbf{r})$ is the eigenfunction in the spinor form, and E is the eigenenergy. $V_0^{(n)}(\mathbf{r})$ is the potential profile across the trilayer: $V_0^{(1)} = V_0^{(3)} = 0$ and $V_0^{(2)} = U_b$. $V_{\rm ex}^{(n)}(\mathbf{r})$ is the exchange splitting potential: $V_{\rm ex}^{(1)} = \Delta_{\rm ex}\sigma_z$, $V_{\rm ex}^{(2)} = 0$, and $V_{\rm ex}^{(3)} = \Delta_{\rm ex}(\sigma_x \sin\theta + \sigma_z \cos\theta)$, where σ_x, σ_y, and σ_z are the Pauli matrices. $V_i(\mathbf{r}) = -U_i\delta(\mathbf{r} - \mathbf{r}_i)$ is the impurity potential, $\mathbf{r}_i = (0, r_i, 0)$ being the impurity position vector.

In order to solve the Schrödinger Eq. (1) in the presence of impurity, we find, first, the wave function, $\psi^0(\mathbf{r})$, and the Green's function, $G^0(\mathbf{r}, \mathbf{r}')$, of the trilayer in the absence of impurity ($U_i = 0$) [11]. The impurity-free trilayer is translational invariant in the xz plane, and therefore the $\mathbf{k}_\parallel$ representation can be employed. The real-space representation is, then, obtained using the inverse Fourier transformation by integrating over $\mathbf{k}_\parallel$ up to a cutoff in-plane momentum k_m taken to be of the order of the reciprocal lattice parameter [12]. This regularization procedure eliminates the divergence in $G^0(\mathbf{r}, \mathbf{r})$ and renormalizes the impurity potential. The impurity state energy E_i is, then, uniquely defined by U_i and k_m. All the results below are presented in terms of the impurity energy E_i, which make them robust to the choice of k_m.

The solution of Eq. (1) can be written in the integral form in terms of $\psi^0(\mathbf{r})$ and $G^0(\mathbf{r}, \mathbf{r}')$ as follows:

$$\psi(\mathbf{r}) = \psi^0(\mathbf{r}) + \int G^0(\mathbf{r}, \mathbf{r}')V_i(\mathbf{r}')\psi(\mathbf{r}')d\mathbf{r}'. \quad (2)$$

Because of the assumed delta potential for impurity, this integral equation can easily be solved:

$$\psi(\mathbf{r}) = \psi^0(\mathbf{r}) - U_i\frac{G^0(\mathbf{r}, \mathbf{r}_i)}{1 + U_iG^0(\mathbf{r}_i, \mathbf{r}_i)}\psi^0(\mathbf{r}_i). \quad (3)$$

We evaluate the IEC using the approach based on the torque produced by rotation of the magnetization of one ferromagnet relative to that of the other [1,13]. The torque is related to spin current j_S in such a way that allows obtaining the IEC constant J from the relationship $J\sin\theta = -\frac{1}{2}\hbar j_S$. Taking into account that the y axis lies perpendicular to the planes, the spin current can be written as follows:

$$j_S = \mathrm{Re}\langle\psi^\dagger\sigma_y v_y\psi\rangle, \quad (4)$$

where ψ is the wave function given by Eq. (3) and v_y is the y component of the velocity operator $\mathbf{v} = -i\hbar\nabla/m$. The angular brackets in Eq. (4) denote averaging over orbital and spin states which involves the integration over the transverse momenta $\mathbf{k}_\parallel$ and the energy E weighted with the Fermi distribution function and the summation over contributions from majority and minority spin electrons incident from the left and right FM layers.

Below we discuss the results of calculations of the IEC. In these calculations we use the parameters characterizing the electronic structure of Fe: $E_F = 2.6$ eV and $\Delta_{\rm ex} = 3.6$ eV, which have previously been used to describe both spin-dependent tunneling [14] and IEC [5]. The volume concentration of impurities is taken to be $n_i = 1.25 \times 10^{21}$ cm^{-3}, which corresponds to about 1% impurity atoms for an insulator of 2 Å interatomic distance. The barrier height is assumed to be $U_b = 1$ eV. This value as well as all the energies below is given with respect to the Fermi energy.

Figure 1 shows the IEC as a function of the impurity energy for different impurity positions r_i within the barrier of thickness $d = 8$ Å. The striking feature evident from this figure is that the IEC has a pronounced peak of the AF exchange for impurity levels lying close to the Fermi energy. Although the width and the amplitude of this peak are sensitive to the impurity position, the coupling constant J is always negative when E_i matches E_F. The absolute values of the IEC are significantly higher than the value of the coupling constant in the same system without impurities which is $J_0 = 1.6 \times 10^{-3}$ ergs/cm^2.

The origin of the AF exchange coupling can be understood from energy-resolved contributions to the spin current from majority and minority spin electrons shown in Fig. 2(a) for $E_i = -0.3$ eV and in Fig. 2(b) for $E_i = 0$. For simplicity only contributions from electrons incident from

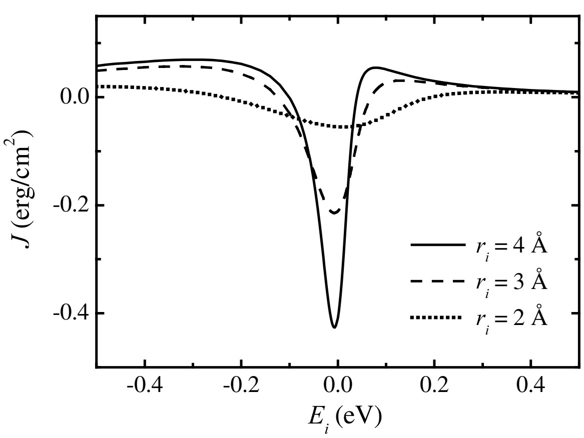

FIG. 1. IEC versus impurity energy E_i (given with respect to E_F) for different impurity positions r_i within the barrier of thickness $d = 8$ Å.

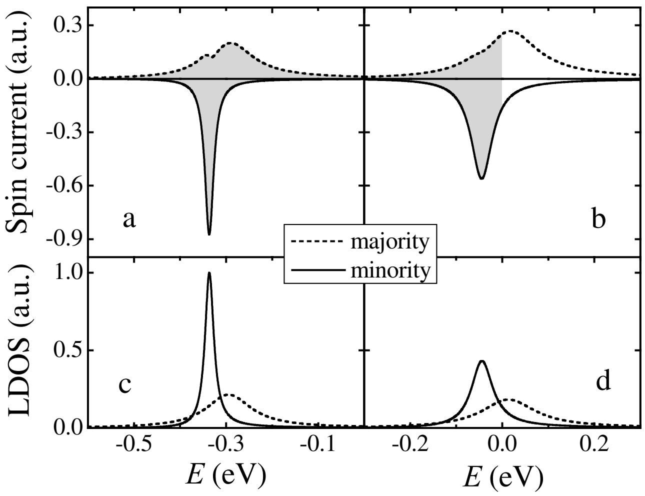

FIG. 2. Energy-resolved contributions to the spin current from majority and minority spin electrons incident from the left ferromagnet [(a),(b)] and local densities of states (LDOS) at the impurity position $\mathbf{r}_i$ [(c),(d)] for $E_i = -0.3$ eV [(a),(c)] and $E_i = 0$ [(b),(d)]. Shadow regions show filled states which contribute to the spin current at $T = 0$, $d = 8$ Å, and $r_i = 3$ Å.

the left ferromagnet are presented. The spin current has a pronounced resonant character reflecting the presence of a spin-split localized state. The latter fact follows from the correlation between the spin current and the local density of states (LDOS) at the impurity position $\mathbf{r}_i$ shown in Figs. 2(c) and 2(d). The majority and minority LDOS peaks are shifted from the impurity energy E_i and have unequal widths due to the coupling to spin-dependent electronic states of the FM metals. The width of the impurity levels is determined by the density of metal induced gap states in a position of impurity, which is larger for majority spins. This is consistent with the first-principles calculation indicating that the majority spin state of Δ_1 symmetry in Fe/MgO/Fe is coupled most effectively from Fe into MgO and decays slowest in the barrier [15]. We note that the shift of the majority and minority levels has opposite sign with respect to the unperturbed impurity level reflecting a different position of this level with respect to the bottom of spin bands.

As is seen from Figs. 2(a) and 2(b), the spin currents have opposite signs (directions) for the majority and minority spins: the torque due to the majority spin current contributes to the FM exchange, whereas the torque due to the minority spin current contributes to the AF exchange. The net spin current (at $T = 0$) is the integral over all the energies up to the Fermi level and the sum over the two spin contributions. If the impurity level lies well below the Fermi energy, as is the case in Fig. 2(a), the net spin current appears to be majority dominated resulting in a FM exchange coupling. However, when the impurity level lies near the Fermi energy, as is the case in Fig. 2(b), the minority spin current exceeds the majority spin current due to the incomplete contribution from the resonant peaks of different width (the shadow regions in Fig. 2(b)] leading to an AF exchange coupling. A particular shape of the resulting AF peak shown in Fig. 1 is also determined by the relative shift of the majority and minority levels shown in Fig. 2.

The energy interval for the AF exchange is determined by the width of the spin-split localized state. This fact explains the variation in the width and in the amplitude of the AF peak with the impurity position shown in Fig. 1. The closer the impurity is placed to the interface, the stronger is the coupling of the impurity level to the electronic states of the ferromagnet and, hence, the broader is the resonant peak. We note an analogy between the impurity-induced AF IEC and the resonant inversion of TMR [16].

In practice impurities (defects) might be distributed randomly within the barrier layer. We have, therefore, averaged the IEC over impurity positions keeping the impurity energy and the volume concentration of impurities fixed. Figure 3(a) shows that the averaged IEC has a pronounced AF peak for impurity levels lying in the vicinity of the Fermi energy. The amplitude and the width of this peak depend on the barrier thickness d, both decreasing with d. As is seen from Fig. 3(b), the variation of the IEC constant as a function of barrier thickness is different depending on the impurity energy. Typically the absolute value of J decays exponentially with d, as is the case for $E_i = 0$ (AF coupling) and for $E_i = 0.2$ eV (FM coupling). However, for certain values of E_i the IEC displays a crossover from AF to FM coupling with increasing d, as occurs for $E_i = -0.2$ eV in Fig. 3(b). A similar behavior of the IEC was found by Faure-Vincent *et al.* [5], but was interpreted as the consequence of the magnetostatic coupling. We note that the first-principles calculation shows a different decay length for bands of different symmetry in Fe/MgO/Fe junctions [15]. Taking into account this behavior should enhance FM coupling at large MgO thickness, because the torque due to the majority spin current contributing to FM exchange will decay slower as the

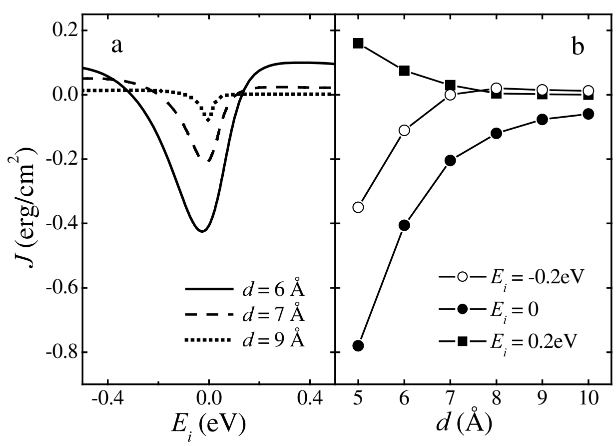

FIG. 3. IEC averaged over impurity position versus (a) impurity energy for different barrier thicknesses and (b) barrier thicknesses for different impurity energies.

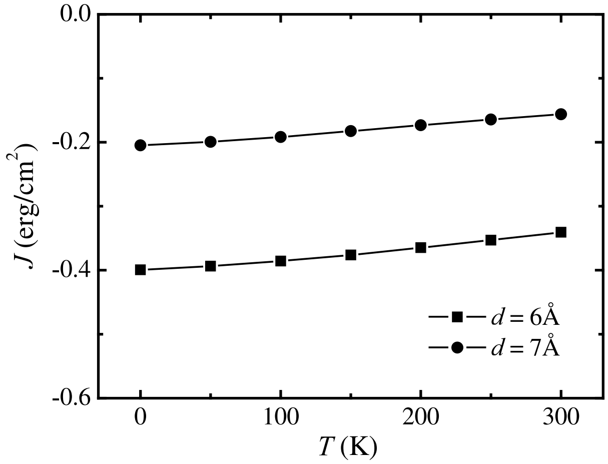

FIG. 4. IEC averaged over impurity position as a function of temperature for different barrier layer thicknesses and $E_i = 0$.

consequence of a slower decay of the majority spin Δ_1 state in MgO.

The temperature dependence of the IEC stems from the thermal broadening of the Fermi distribution. Because of the resonant character of the impurity-assisted coupling, this broadening smears out the energy dependence of the spin current convoluted with the Fermi distribution function [Fig. 2(b)]. Therefore, with increasing temperature the IEC constant becomes a smoother function of the impurity energy. This leads to a monotonic decrease in the AF exchange with temperature, as is seen in Fig. 4 for $E_i = 0$. This result is opposite to that predicted for a perfect barrier, for which the thermal population of the electronic states above the Fermi level leads to an increase in the IEC with temperature [2], but is consistent with the experimental finding [7].

In order to elucidate the strong AF coupling observed in Fe/Si/Fe(001) junctions [6,7] we repeated the calculation assuming a barrier height of 0.3 eV, appropriate for Si [7]. We found that experimentally measured AF coupling of 2 ergs/cm^2 can be obtained within our model assuming homogeneous distribution of impurities placed at the Fermi energy for barrier thickness $d = 5$ Å and impurity concentration, n_i, of about 1 at. %. The same magnitude of J is obtained for $d = 10$ Å and $n_i \approx 12\%$. This is consistent with the known fact of strong interdiffusion between Si and Fe which might either reduce the effective barrier thickness or create a large concentration of Fe impurities in Si [6].

In conclusion, we have shown that the presence of impurity or defect states in the insulating barrier layer separating two ferromagnets affects dramatically the interlayer exchange coupling. The resonant origin of the impurity-assisted IEC makes the coupling much stronger than that in the absence of impurities. The IEC becomes antiferromagnetic and decreases with temperature if the energy of the impurity state matches the Fermi energy. The predicted AF exchange coupling might be typical for epitaxial tunnel junctions if the sufficient number of impurity or defect states is created during the growth. A particular configuration of these localized states with a well-defined energy in the band gap of the insulator may pin the Fermi level to this energy [17]. This elucidates the available experimental results on the antiferromagnetic IEC in epitaxial tunnel junctions.

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