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# Effect of oxygen vacancies on interlayer exchange coupling in Fe/MgO/Fe tunnel junctions

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**Abstract:** We have investigated the interlayer exchange coupling (IEC) in Fe/MgO/Fe(0 0 1) tunnel junctions with and without oxygen vacancies in MgO, using model and density functional calculations. The model predicts that IEC changes sign from ferromagnetic to antiferromagnetic if a defect level matches the Fermi energy. Ab initio calculations show that for perfect junctions, IEC is ferromagnetic and decreases exponentially with MgO thickness. Oxygen vacancies placed in the middle of MgO make IEC antiferromagnetic for three monolayers (MLs) of MgO, but do not change the sign of IEC for five MLs. The latter fact is explained within the model, which suggests that for the impurity level lying below the Fermi energy IEC can change sign with increasing barrier thickness due to the weaker coupling of the impurity level to the ferromagnets.

**Keywords:** Interlayer exchange coupling, Impurity, O vacancy, Magnetic tunnel junction, Spin current, Ab initio calculations

## 1. Introduction

Interlayer exchange coupling (IEC) takes place in magnetic multilayers in which ferromagnetic layers are separated by a nonmagnetic spacer. The spacer can be conducting as well as insulating. In the case of a conducting spacer, the interlayer coupling has an oscillating character, similar to RKKY–interaction, as a function of the spacer thickness. An insulating spacer leads to exponentially decaying non-oscillatory dependence of the strength of IEC (for a recent review, see Ref. [1]).

There are several theoretical approaches to calculate IEC. The first study of IEC in magnetic tunnel junctions (MTJs) was performed by Slonczewski [2] who used a method based on spin current. An approach based on quantum interference due to spin-dependent reflections at the interfaces was used by Bruno [3] and [4] and Stiles [5]. Another way to calculate IEC is to use ab initio calculations of the ground state energy of the MTJ for parallel and antiparallel configuration of the mag-

netization. It follows from the theoretical considerations [2], [3], [4] and [5] that a change in the electronic density of states has a crucial importance for the strength of IEC. Therefore, any significant change in electron wave functions leads to a sizable change of IEC.

Experimental observation of IEC across a tunneling barrier is much more demanding compared to metallic spacers. The reason is that producing a high-quality MTJ with a thin barrier is a very complicated task. There are only few reports of measurements of IEC in MTJs [6], [7] and [8]. The results obtained in these experiments significantly differ from the theoretical predictions [2], [3], [4] and [5], both in the magnitude and the sign of IEC.

We have shown in our previous paper [9] that this discrepancy may be attributed to the presence of impurities or defects in the barrier layer. We found that the resonant origin of the impurity-assisted IEC makes the coupling much stronger than that in the absence of impurities. IEC becomes antiferromagnetic and decreases with temperature if the energy of the im-

purity state matches the Fermi energy. These results are consistent with the experimental observations [7] and [8].

In this paper, we consider IEC in Fe/MgO/Fe junctions. A well-known type of defects in MgO are O vacancies which create localized states in the band gap of MgO. Using parameters suitable for Fe/MgO/Fe junctions we perform model calculations and analyze the influence of O vacancies on IEC. We carry out first-principles calculations and show that O vacancies can change the sign of IEC, confirming our model results.

## 2. Model calculation of IEC

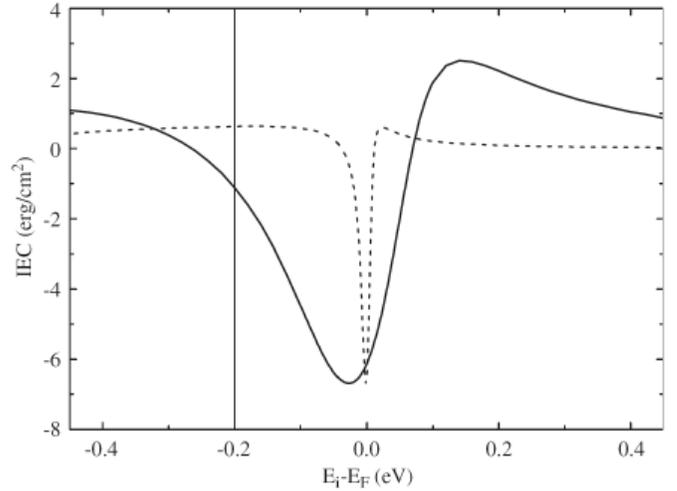
To calculate IEC in Fe/MgO/Fe MTJs we follow the Slonzewski's approach [2], presented in details in Ref. [10]. According to this approach, for arbitrary angle  $\theta$  between magnetic moments of the two ferromagnetic layers the time rate of change of the relevant component of the expectation value of electron spin  $\langle \dot{S}_y \rangle$  is proportional to the spin current,  $j_y$ , which follows from the continuity equation ( $y$ -axis is chosen to be perpendicular to the layer of the MTJ). On the other hand,  $\langle \dot{S}_y \rangle$  represents the torque which is equal to the derivative of the exchange energy with respect to the angle  $\theta$ . Therefore, the IEC strength,  $J$ , can be found from the relationship  $J \sin \theta = -1/2 \hbar j_y$ .

We use a free-electron-like model to describe exchange-split conduction bands of the ferromagnets. An impurity is modeled by a delta potential  $V_i(\mathbf{r}) = -U_i \delta(\mathbf{r} - \mathbf{r}_i)$ , where  $\mathbf{r}_i$  is the position of impurity level. An insulating barrier is described using a rectangular potential of height  $U_b$ . The single-particle wave function  $\psi(\mathbf{r})$  in the  $n$ th layer obeys the Schrödinger equation

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

Here,  $n$  is the layer index— $n = 1$  and  $3$  denote the left and right ferromagnets, and  $n = 2$  denotes the barrier.  $V_0^{(n)}(\mathbf{r})$  is the potential profile across the trilayer— $V_0^{(1)} = V_0^{(3)} = 0$  and  $V_0^{(2)} = U_b$ .  $V_{\text{ex}}^{(n)}(\mathbf{r})$  is the exchange splitting potential— $V_{\text{ex}}^{(1)} = \Delta_{\text{ex}} \sigma_z$ ,  $V_{\text{ex}}^{(2)} = 0$ , and  $V_{\text{ex}}^{(3)} = \Delta_{\text{ex}} (\sigma_x \sin \theta + \sigma_z \cos \theta)$ , where  $\sigma_x$ ,  $\sigma_y$  and  $\sigma_z$  are the Pauli matrices. The wave function of the system with impurity can be expressed through the wave function of the perfect system,  $\psi^{(0)}(\mathbf{r})$ , and its Green function [9]. The latter is obtained using a method developed in Refs. [11] and [12]. The calculated wave function is used to obtain the spin current,  $j_y$ , which is related to IEC, as was described above.

In our calculations of IEC in Fe/MgO/Fe junctions, we used the following parameters to describe Fe:  $E_F = 2.6$  eV,  $\Delta_{\text{ex}} = 3.6$  eV. The MgO barrier height,  $U_b = 1.16$  eV, was obtained from ab initio calculations (see Section 3). Results are presented in Figure 1 which shows IEC as a function of impurity energy,  $E_i$ , for two barrier thicknesses  $d = 6$  and  $10$  Å. Here, it is assumed that the impurities lie in the middle of the barrier and have an in-plane concentration  $n = 1/32$  Å<sup>-2</sup> (which



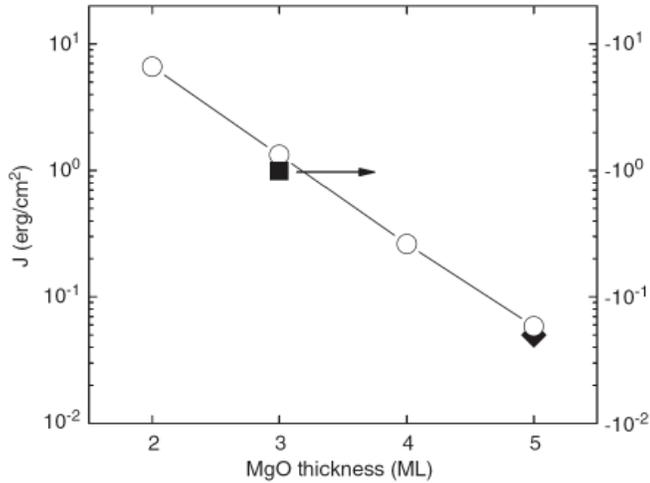
**Figure 1.** IEC versus energy,  $E_i$ , of impurities placed in the middle of the barrier of thickness  $d = 6$  Å (solid line) and  $d = 10$  Å (dashed line). The latter is multiplied by a factor of 10. The in-plane concentration of impurities is  $n = 1/32$  Å<sup>-2</sup>.

corresponds to one impurity per eight host atoms in the plane, assuming that the lattice parameter  $a = 2$  Å). IEC has a pronounced peak of the antiferromagnetic exchange for impurity levels lying close to the Fermi energy. The origin of the impurity-induced antiferromagnetic IEC has similarity to the resonant inversion of tunneling magnetoresistance [13] and [14] and was explained in Ref. [9].

It is seen from Figure 1 that the amplitude and the width of this antiferromagnetic peak are very sensitive to the barrier thickness. Both decrease exponentially with the increase of  $d$ , reflecting the exponentially weaker coupling of the impurity level to the ferromagnets (note that in Figure 1  $J$  is enhanced by a factor of 10 for  $d = 10$  Å). For impurity energies away from the resonance, IEC becomes ferromagnetic. The scale of this change is controlled by the width of the peak. Since the latter decreases with  $d$ , there is a range of impurity energies at which the coupling can change sign. For example, for  $E_i = E_F - 0.2$  in Figure 1 (shown by the vertical line), IEC is negative for  $d = 6$  Å, but positive for  $d = 10$  Å. Such a crossover from antiferromagnetic to ferromagnetic coupling was observed by Faure-Vincent et al. [7] but was interpreted as a consequence of the magnetostatic coupling.

## 3. Ab initio calculation of IEC

Ab initio calculations of the interlayer exchange coupling in Fe/MgO/Fe(001) junctions have been performed using a pseudopotential plane-wave method implemented in the Vienna Ab-initio Simulation Package (VASP) [15], [16] and [17]. Since only periodic systems can be studied in VASP, we considered two Fe slabs of five monolayers (MLs) each, separated by two MgO slabs of  $N$  MLs each. The interlayer distances were taken as follows:  $a_{\text{Fe}} = 2.87$  Å,  $a_{\text{MgO}} = \sqrt{2}a_{\text{Fe}}$  Å and  $a_{\text{Fe-O}} = 2.17$  Å [18]. The total energy of the system was

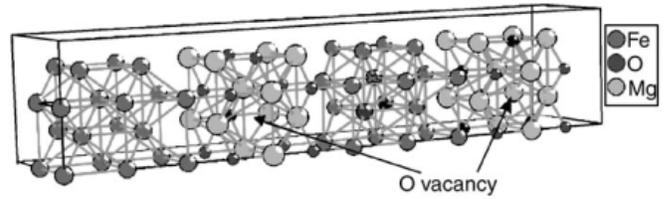


**Figure 2.** IEC as a function of barrier thickness for a perfect Fe/MgO/Fe trilayer (open symbols) and for the same system with oxygen vacancies in MgO (solid symbols). The ■ point is plotted against the negative scale to the right.

calculated for parallel and antiparallel magnetizations of the Fe layers,  $E_p$  and  $E_{AP}$ , respectively. The magnitude of IEC was obtained from  $J = (E_{AP} - E_p)/2$ . Self-consistent calculations were performed using 21 k-points in the irreducible surface Brillouin zone and the energy was converged to  $10^{-9}$  eV.

The epitaxial Fe/MgO/Fe junction with no vacancies shows ferromagnetic IEC which decreases exponentially with the MgO thickness,  $N$ , as is shown in Figure 2. It is interesting that the calculated magnitude of IEC is very large. Even for 5MLs of MgO ( $d \approx 1$  nm) we find  $J \approx 0.06$  erg/cm<sup>2</sup>, which is comparable to values measured for some metallic spacer layers. Taking into account that  $J \propto e^{-2\kappa d}$  [2], we obtain the decay constant  $\kappa = 1.1 \text{ \AA}^{-1}$ . This allows us to estimate the effective potential barrier height of  $U_b = 1.16$  eV, which was used in our model calculations.

The Fe/MgO/Fe junction with O vacancies in the middle of the MgO layer is illustrated in Figure 3. Atomic structure relaxation shows no significant distortion of the crystal structure. We find that the vacancy introduces an impurity band lying about 1 eV below the Fermi level. This band has a width of the order of 1 eV due to the relatively large concentration of vacancies we used in our calculations. The effect of O vacancies in MgO on IEC is also shown in Figure 2. For 3MLs of MgO we find that the presence of O vacancies reverses IEC from ferromagnetic to antiferromagnetic. The magnitude of the antiferromagnetic coupling is  $J = -0.99$  erg/cm<sup>2</sup>. For 5MLs of MgO the coupling becomes ferromagnetic,  $J = 0.055$  erg/cm<sup>2</sup>, which is slightly less than the IEC value for the system without vacancies. The reversal is consistent with the experimental observations [7] and may be explained using the arguments of our simple model presented in Section 2. Since the Fermi level lies above the vacancy level, we can expect that the width of the antiferromagnetic peak decreases with MgO thickness which may lead to the change of sign of IEC.



**Figure 3.** Unit cell of the Fe/MgO multilayer used in total energy calculations.

## 4. Conclusions

We have investigated the interlayer exchange coupling in Fe/MgO/Fe(0 0 1) junctions which contain O vacancies in the MgO barrier using the model and ab initio calculations. The model calculations predict that IEC changes sign from ferromagnetic to antiferromagnetic if an impurity (defect) level matches the Fermi energy. Ab initio calculations show that for perfect junctions IEC is ferromagnetic, and decreases exponentially with the barrier thickness. Oxygen vacancies placed in the middle of MgO make IEC antiferromagnetic for 3MLs of Mg, but do not change the sign of IEC for 5MLs. The latter fact is explained within the model, which suggests that for the impurity level lying below the Fermi energy, IEC can change sign with increasing barrier thickness due to the weaker coupling of the impurity level to the ferromagnets.

## Acknowledgments

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