Tunneling interlayer exchange coupling between oxide ferrimagnets: analysis for Fe_3O_4/vac/Fe_3O_4 case

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Abstract

We have investigated tunneling interlayer exchange coupling (TIEC) between ferrimagnetic Fe_3O_4 films via a tunneling barrier. In this investigation we employ *ab-initio* density functional theory to study a generic tunneling junction incorporating ferrimagnets Fe_3O_4/vac/Fe_3O_4. In contrast with previously established TIEC theory calculated here thickness dependence is non-monotonic and accompanied by TIEC sign change. Our calculations clearly demonstrate that TIEC is controlled mainly by an interfacial oxygen induced spin polarization. These results emphasize importance of localized and strongly directional electronic interactions at the interface, thus showing limitations of free electron model treatment of the problem.

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Magnetic tunneling junctions (MTJs) are active area of research. This is[1] due to both interesting fundamental physics as well as significant technological potential for these devices[2]. The latter have been recently demonstrated with FeCoB/MgO junctions which are now considered as the industry standard for hard disk drives (HDD) . After these recent successes, MTJ's are currently studied as possible critical elements of emerging spin electronic devices such as spin torque magnetic random access memory (ST-MRAM)[3].

Using ST-MRAM as one specific example, a critical issue in making this storage technology commercially competitive is scalability, or the ability to make ever smaller diameter current-perpendicular-to-plane (CPP) devices based on MTJs while keeping excellent demonstrated properties intact. One of the fundamental issues associated with this is the dipolar magnetic field of neighboring devices[4]. This and other fundamental issues continue to motivate the search for magnetic materials to be incorporated into MTJs in order to circumvent critical fundamental technology issues. The use of ferrimagnetic materials with a reduced saturation magnetization while still showing significant magnetoresistance (MR) properties is seen as one of the attractive pathways. Indeed, Fe_3O_4 is one of such materials since it is one of the few conducting, ferrimagnetic and so-called half-metallic oxides[5].

The tunneling interlayer exchange coupling (TIEC) across an insulating nonmagnetic spacer, therefore, has also attracted renewed attention recently. This phenomenon is instrumental in the context of various spintronic effects based on MTJs or all metal giant magnetoresistance (GMR) heterostructures[6,7]. Recently, a strong antiferromagnetic (AF) coupling in a fully epitaxial Fe/MgO/Fe trilayer structure has been observed by several groups[8,9]. Various theoretical models have been proposed to explain the origin of TIEC. Some of these models explain the AF TIEC as the result of impurities[10] or vacancies[11] in the tunneling barrier. Despite all of the intensive research over the last decade, details of the tunneling TIEC between ferromagnetic films via a tunneling barrier is still under debate[12]. The TIEC between ferrimagnetic films is much less investigated and understood, despite the scientific interest and its potential technological importance.

In this Letter, we report the results for TIEC between Fe_3O_4 ferrimagnets. Since, the best, in terms of TMR tunneling barrier for this material is yet to be discovered, we consider here the case of Fe_3O_4/vac/Fe_3O_4 MTJ. We employ density functional theory (DFT) to describe this hetero-structure as it is well established that local-spin-density-approximation (LSDA) reproduces essential properties of Fe_3O_4 for this study, such as ferrimagnetism, half-

metalicity, and provides a reasonable estimate of the critical temperature[13]. However, as expected it fails to describe charge ordering states[14]. Most interestingly, the DTF-LSDA calculations uncover the importance of the spin polarized interface oxygen states on the sign of TIEC.

To investigate the TIEC between ferrimagnetic Fe_3O_4 films, we performed *ab-initio* calculations for the Fe_3O_4/vac/Fe_3O_4 system. We choose the VAC barrier as an example of a generic tunneling barrier with an intention to identify features of TIEC between ferrimagnetic oxide films. We employed the projected augmented plane wave method (PAW)[15], implemented within the Vienna *Ab Initio* Simulation Package (VASP)[16]. The valence configuration $3d^74s^1$ for Fe and $2s^22p^4$ for O were used. The generalized gradient approximation[17] is used for the exchange-correlation energy. In our calculations, we consider two reduced Fe_3O_4 unit cells of 28 atoms each, separated by a vacuum spacer to form periodic computation cell shown in Fig. 1. The orientation of Fe_3O_4 is (100) along the barrier surface which is consistent with experimental conditions in[18]. TIEC with different K-points was calculated until reaching the convergence of 0.1 meV. For 0.42 nm vacuum spacer, $a = b = 5.93364\mathring{A}$, $c = 25.1828\mathring{A}$ and a monkhorst-pack 16×16×4 k-point grid were applied. Note, that we have kept $N_x/N_z = c/a$ for all the calculation, where N_x (N_z) is the number of k-points.

To determine the sign and strength of TIEC, we calculated from first principles the total energy difference between parallel (E_P) and antiparallel (E_{AP}) magnetization configurations given generally by

$$E_{tot} = \int_{-\infty}^{\epsilon_F} d\epsilon (N^{\uparrow}(\epsilon) + N^{\downarrow}(\epsilon))(\epsilon - \epsilon_F) \; - E_{dc} + E_{es}, \tag{1}$$

where the first terms is for band energy expressed through the density of states for spin up and down ($N^{\downarrow}(\epsilon)$); second for so-called double counting and third for electrostatic energy. This total energy difference can be mapped onto the Heisenberg model where the energy is determined as

$$E = -N * J_1 cos\theta, \tag{2}$$

where θ is the angle between the magnetization directions of two adjacent Fe_3O_4 layers and N is the number of nearest neighbors and J_1 is TIEC constant. Here J_1 signifies the effective long-range bilinear TIEC constant. The Positive sign of the J_1 constant would correspond to a ferromagnetic TIEC favoring the parallel alignment of the Fe_3O_4 layer magnetization.

The magnitude of TIEC strength J_1 was obtained from $J_1 = (E_{AP} - E_P)/4$ formula accounting for a number of neighbors in the case of using periodic boundary conditions. These calculations are summarized in Fig.2. We found that unexpectedly two Fe_3O_4 layers are antiferromagneticaly coupled through the vacuum barrier when its thickness is between 0.84 nm and 2 nm. Remarkable that this range of thicknesses in good agreement with our early experimental results[18] for TIEC in Fe_3O_4/MgO/Fe_3O_4 MTJs. Interesting also that as the spacer thickness is below 0.84 nm or above 2 nm, the sign of TIEC changes from negative to positive.

In contrast to recently published experiment and theory results for MgO mediated TIEC between ordinary ferromagnetic metals[1,7–11,19,20], we find non-monotonic dependence of TIEC on spacer thickness moreover we find that TIEC changes sign. In earlier calculations[10,11,20], the TIEC is ferromagnetic (ideal junctions) or antiferromagtic (with oxygen vacancies) nature and its TIEC strength decreases exponentially with increasing the barrier thickness. It seems reasonable to expect that the TIEC is very sensitive to the interface states[20]. The calculated electronic structure of the bulk atoms was consistent with previous publication[21]. Here, in this paper, we considered two Fe_3O_4 layers magnetic exchange coupled through the vacuum barrier layer. The electronic structure of the surface atoms is vacuum thickness dependent which is different from the bulk Fe_3O_4 or the Fe_3O_4 surface with thick vacuum slab. In Fig. 3, we plotted the density of states (DOS) of the interface oxygen atoms (marked as 1 in Fig.1) of the second Fe_3O_4 layers for the case of the vacuum thickness of 0.42 nm, 0.84 nm, and 1.68 nm respectively. One can clearly see from Fig. 3 that, for 0.42 nm vacuum spacer, that when the magnetization of two Fe_3O_4 layers changes from antiparallel to parallel configuration, the total DOS of oxygen shifts to the low energy state. In contrast, for 1.68 nm vacuum spacer, when the magnetization of two Fe_3O_4 layers changes from antiparallel to parallel configuration, the total DOS of oxygen shifts to the high energy state which means a strong AF TIEC. One can also see that, for the case of 0.42 nm thick vacuum spacer, the state of oxygen atoms for the parallel configuration is quite different from that for the antiparallel configuration and the shift between the two magnetization configurations is larger than that for 0.84 nm thick vacuum. According to the Eq. 1 term 1, the large shift of the DOS to lower energy would mean larger total energy difference and correspondingly stronger TIEC. We can also see from Fig. 3 that the interface oxygen for all vacuum thicknesses has been strongly spin polarized. Fig. 4 shows

the calculated induced magnetic moment of interface oxygen atoms and the calculated total magnetic moment of the system as a function of the vacuum thickness for the case of the magnetization of two Fe_3O_4 layers in the parallel configuration. Interestingly, the induced magnetic moment and the total magnetic moment follows very closely the calculated TIEC thickness dependence. This is indicative of dominating contribution of the interface oxygen states to TIEC.

The TIEC can be discussed on the basis of the quantum interference theory[20]. It is well-known that the quantum interference is spin-dependent and the reflection coefficient at the interface depends on the direction of the electron spin with respect to the magnetization direction. For the positively polarized magnetization, the TIEC is of FM nature. In contrast, for negative induced magnetization, the reflection coefficient at the interface has been modified and further causes the TIEC sign change. Our results show that the interface oxygen has a high polarizability and a strong non-local effect, especially a strong effect on TIEC. These findings emphasize importance of the strong chemical bonding between interface oxygen and Fe atoms, the characteristic of the system which is not expected to be described within commonly used for modeling TIEC free electron model[10] and references in this pubclication. These findings are believed to be of general importance for the optimization and development of tunneling devices for various spintronic applications.

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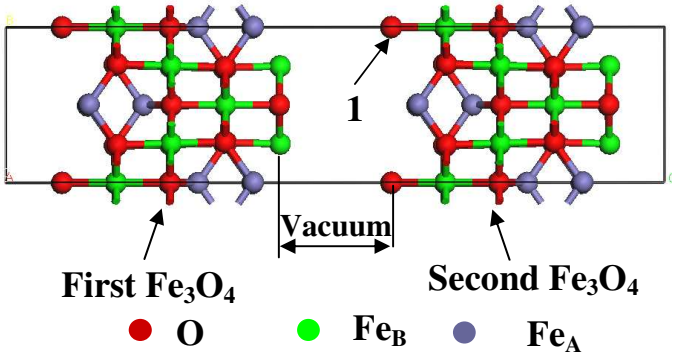

FIG. 1: (Color on line) Schematic of the unit cell of the Fe_3O_4/vac/Fe_3O_4 junction used in total energy calculations.

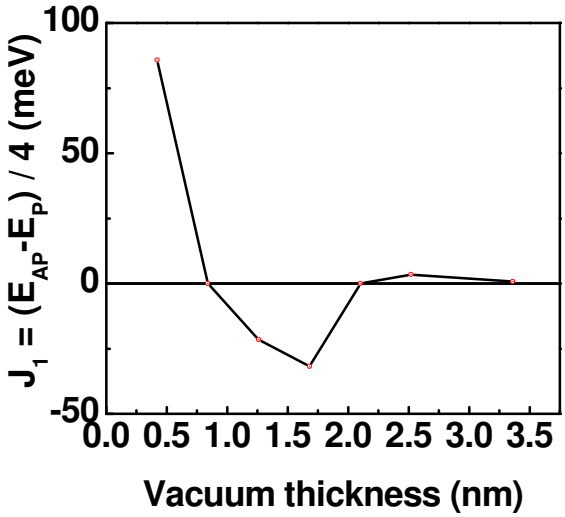

FIG. 2: (Color on line) Calculated TIEC J_1 for Fe_3O_4/vac/Fe_3O_4 junctions as a function of vacuum spacer thickness.

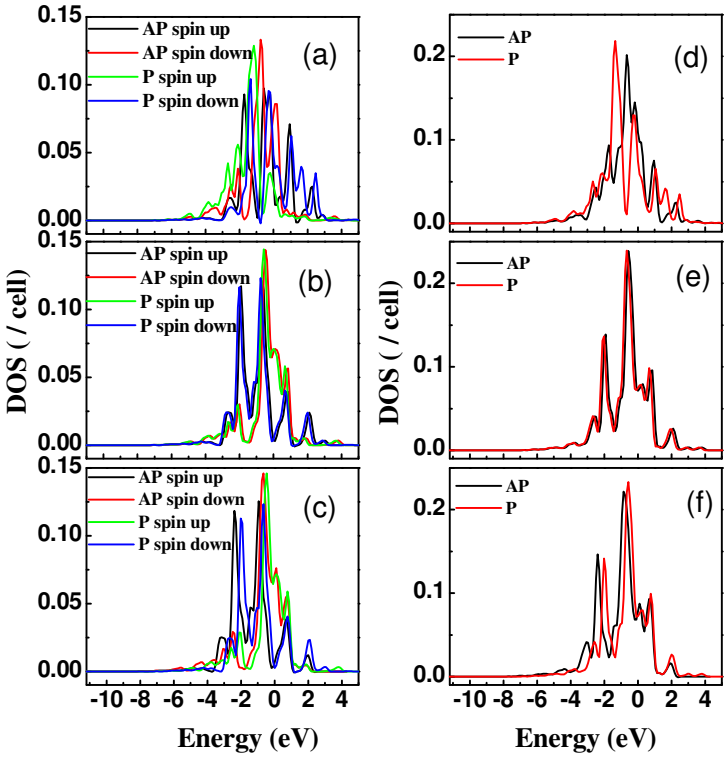

FIG. 3: (Color on line) Calculated DOS of the interface oxygen atoms (marked as 1 in Fig.1) of the second Fe_3O_4 layer for the case of the vacuum thickness of 0.42 nm (a), 0.84 nm (b), and 1.68 nm (c) respectively. Figs. d-f are the calculated total DOS (spin up + spin down) of the interface oxygen atoms for for the case of the vacuum thickness of 0.42 nm, 0.84 nm, and 1.68 nm respectively.

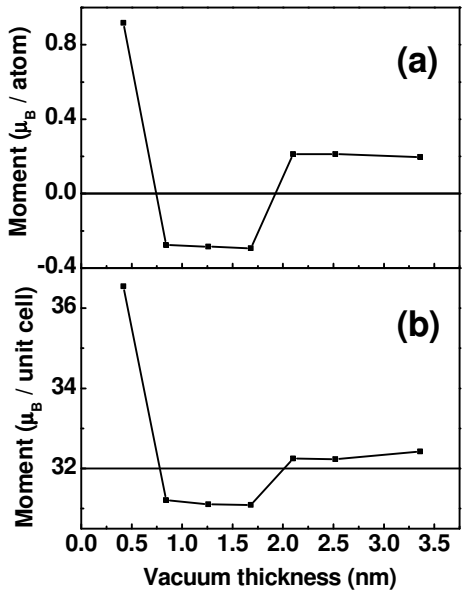

FIG. 4: Calculated induced magnetic moment of interface oxygen atom (a) and the calculated total magnetic moment (b) of the system as a function of the vacuum thickness for the case of the magnetization of two Fe_3O_4 layers in parallel configuration.