Effect of structural relaxation and oxidation conditions on interlayer exchange coupling in Fe|MgO|Fe tunnel junctions

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The effect of structural relaxation and interfacial oxidation is demonstrated from first principles to have a crucial impact on interlayer exchange coupling (IEC) in crystalline Fe|MgO|Fe magnetic tunnel junctions (MTJs). It is shown that the IEC becomes antiferromagnetic for the relaxed structure in contrast to ferromagnetic for unrelaxed Fe|MgO|Fe MTJs. Furthermore, the antiferromagnetic IEC is strongly enhanced in the presence of oxygen vacancies while it is decreased by overoxidation and may even become ferromagnetic for sufficiently high oxygen concentration at the Fe|MgO interface. These results were supported using a tight-binding model and provide an explanation for recent experimental studies. © *2010 American Institute of Physics*. [doi:10.1063/1.3459148]

Since its observation in Fe|Cr|Fe layered structures,[1] interlayer exchange coupling (IEC) has been a subject of major interest to the spintronics community, both from fundamental and applications points of view.[2] This interest was strongly enhanced when antiferromagnetic (AF) IEC between ferromagnetic (FM) layers across an insulator was found in Fe|MgO|Fe crystalline magnetic tunnel junctions (MTJs).[3] Experimentally observed values for the IEC may reach up to -0.26 erg/cm^2 for thin MgO layer thicknesses.[3,4] The AF IEC phenomenon was initially explained using the free electron model for pure tunnelling[3,5] and the resonant tunneling mechanism due to the presence of localized impurity or defect states in the insulating barrier layer.[6,7] For instance, it was shown from first principles that oxygen vacancies located in the middle of the MgO layer[4,7] as well as interfacial oxygen[8] cause the IEC to be AF for MgO thicknesses below 0.8 nm. In a recent experiment, Chiang *et al.*[9] investigated the role of interfacial oxidation on the IEC in Fe|MgO|Fe MTJs and found that oxidation conditions can strongly affect the character of the IEC for MgO thicknesses below 1 nm. They observed that for MgO thicknesses up to 0.6 nm both under- and overoxidized junctions show AF IEC which is much stronger in case of underoxidation. However, the IEC becomes FM in case of overoxidized samples for MgO thicknesses larger than 0.6 nm [~3 monolayers(ML)].

Motivated by these observations and desiring to elucidate the impact of oxidation conditions on the nature of the IEC in single crystal MTJs, we performed systematic *ab initio* studies of (i) the influence of oxygen impurities and vacancies and (ii) the effects of structural relaxation on the IEC. We found that oxygen vacancies cause strong AF IEC in agreement with previous theoretical[4,7] and experimental[4,9] studies. Furthermore, interfacial oxidation decreases the AF IEC and may even lead to FM IEC depending on the interfacial oxygen concentration. Most importantly, we theoretically demonstrate that the full structural relaxation of pure Fe|MgO|Fe MTJs may lead to AF IEC.

We carried out first-principles calculations by using the Vienna *ab initio* simulation package (VASP).[10–12] In our calculations, the electron-core interactions were described by the projector augmented wave method for the pseudopotentials,[13] and the exchange correlation energy was obtained within the generalized gradient approximation (GGA).[14] The cutoff energies for the plane wave basis set used to expand the Kohn-Sham orbitals were 400 eV for the ionic relaxations and 500 eV for the subsequent self-consistent FM (AF) energy calculations.

For purposes of the present work, we constructed periodic supercells of Fe|MgO|Fe|MgO comprising thicknesses between 2 and 5 MLs for MgO layers with the thickness of

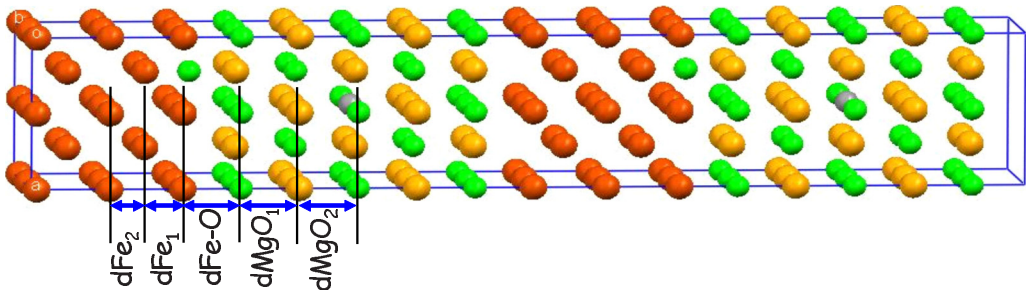

FIG. 1. (Color online) 2×2 supercell of $[Fe_5(MgO)_5]_2$ used for the calculations of additional oxygen located at the Fe|MgO interface and oxygen vacancy in the middle of MgO layer.

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TABLE I. Interlayer distances (Å) of $[Fe_5(MgO)_x]_2$. Unrelaxed results are taken from Ref. 15, the values of $Fe_{10}(MgO)_6$ are taken from Ref. 16.

Structures	dFe_1	dFe_2	dFe-O	$dMgO_1$	$dMgO_2$
$[Fe_5(MgO)_2]_2$	1.252	1.405	2.115	2.149	
$[Fe_5(MgO)_3]_2$	1.240	1.398	2.101	2.099	2.099
$[Fe_5(MgO)_4]_2$	1.236	1.395	2.097	2.099	2.120
$[Fe_5(MgO)_5]_2$	1.231	1.392	2.092	2.094	2.111
Unrelaxed	1.433	1.433	2.160	2.026	2.026
$Fe_{10}(MgO)_6$(GGA)	1.350	1.427	2.219	2.219	2.177
$Fe_{10}(MgO)_6$(LSDA)	1.120	1.343	2.002	2.130	2.119

the Fe layers kept equal to 5 MLs. The lateral cell size accommodated 4 Fe, 4 Mg, and 4 O per layer or 8 Fe, 8 Mg, and 8 O per layer [Fig. 1]. The Monkhorst–Pack scheme was used for the Brillouin zone integration. A $21\times21\times1$ k-point mesh was sufficient to ensure good convergence in the total energy differences. Full structural relaxations in shape and volume were performed ensuring that the Hellmann–Feynman forces acting on ions were less than 10^{-3} eV/Å.

The IEC constant J across one barrier region and normalized by the lateral area of the computational cell is defined as

$$J=(E_{AP}-E_P)/2, \tag{1}$$

where E_P and E_{AP} represent total energies for parallel and antiparallel alignments of adjacent Fe layer magnetizations, respectively. To ensure an accurate value for J, calculations were performed in three steps. First, we relaxed the structure with parallel alignment of Fe slabs magnetizations. Next, we calculated the total energy $E_P(E_{AP})$ for FM (AF) configuration with the relaxed structure. Finally, the strength of the IEC was calculated using formula (1).

We begin by describing the results of the structural relaxation of the "pure" structure, i.e., with no vacancies and/or additional oxygen. The longitudinal interatomic distances along the out-of-plane (001) direction are summarized in Table I. One can note that the interlayer distances within Fe are strongly decreased compared to unrelaxed structures used in Ref. 15. Furthermore, the distances between the first and second interfacial Fe layers (dFe_1) are shorter than the distances between the second and third ones (dFe_2). This is consistent with previous results for $Fe_{10}(MgO)_6$ (Ref. 16) and can be attributed to the formation of the interface. At the same time, the thicknesses of MgO slabs are less affected compared to unrelaxed structures but the MgO interlayer distances are still shorter around the Fe|MgO interface compared to those within the bulk. Finally, the interfacial Fe–O distances are found to be slightly shorter compared to unrelaxed distances (2.16 Å) or bulk FeO (2.154 Å) (Ref. 17) and fall between values obtained within GGA and local spin density (LSDA) approximations in $Fe_{10}(MgO)_6$ (Ref. 16) (see Table I). The discrepancies between aforementioned interlayer distances and those reported in Ref. 16 within GGA approximation are due to full structural relaxation in shape and volume performed in our case unlike that in Ref. 16.

In the lateral direction, the Fe layers expand while the MgO layers shrink compared to the bulk values for bcc Fe (2.866 Å) and rocksalt MgO ($\sqrt{2}/2a_{MgO}$=2.977 Å), respectively, as shown in Fig. 2. One can indeed note that for all cases investigated the relaxed lateral lattice constants are found to be distributed between those for the bulk bcc Fe and MgO (Fig. 2). This result can be viewed as a relaxation "compromise" between these materials originating from the mismatch between different crystal lattices comprising the supercell under investigation.

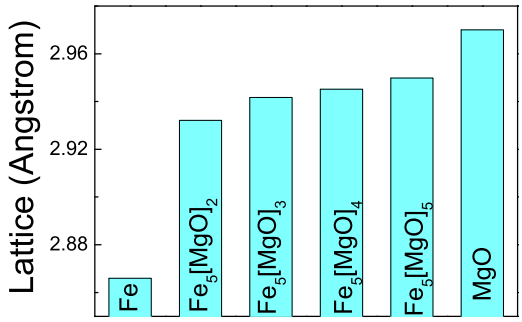

FIG. 2. (Color online) In plane lattice constant of $[Fe_5(MgO)_x]_2$ as a function of MgO thickness.

Let us now to discuss the calculated IEC values for these structures. In Fig. 3(black circles) we show the dependence of the IEC on MgO thicknesses between 2 and 5 MLs. Surprisingly, the IEC is found to be AF unlike the case of unrelaxed structures where we found the IEC to be FM in agreement with Refs. 4 and 7. This underlines the crucial importance of structural relaxation on IEC similarly to the case of transport properties in Fe|MgO MTJs.[16] The values of IEC for relaxed structures can reach up to -0.406 erg/cm^2 and -0.038 erg/cm^2 for structures with 2 and 3 MLs of MgO, respectively. These values are in agreement with experimental results[3] as shown with open squares in Fig. 3. To support these *ab initio* results, we performed total energy calculations using a single-band tight-binding model. This method qualitatively predicts the correct voltage dependences of the spin transfer torques (STT) in MgO-based MTJs.[18–20] Taking the parameters[23] from Refs 19 and 20 corresponding to the measured STT voltage dependences,[21,22] the resulting IEC turns out to be AF in agreement with the *ab initio* results (Fig. 3) providing support for simple modeling approaches to spin-dependent tunneling in MTJs.

Next, we investigated the effects of oxidation conditions on IEC in the Fe|MgO|Fe MTJs. The underoxidation was modeled by creating an oxygen vacancy in the middle of the MgO (Fig. 1) while the overoxidized case was represented by additional oxygen atoms at the Fe|MgO interface (Fig. 1).

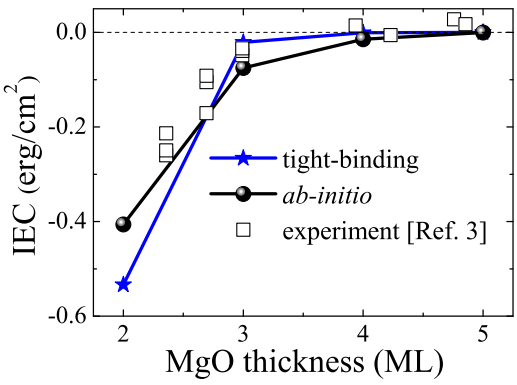

FIG. 3. (Color online) IEC for $[Fe_5(MgO)_x]_2$ calculated using *ab initio* and tight-binding methods together with experimental results taken from Ref. 3.

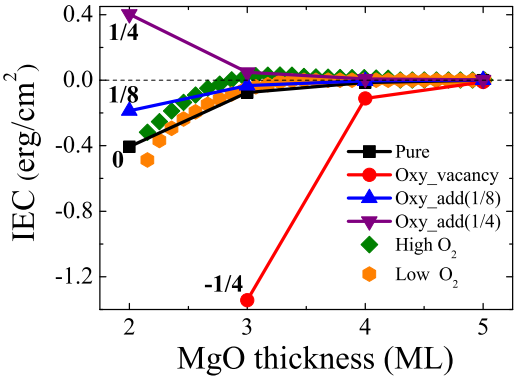

FIG. 4. (Color online) IEC for $[Fe_5(MgO)_x]_2$ pure structure (black squares), for the case with oxygen vacancy in the MgO layer (red circles), and two cases with 1/8 (blue up triangles) and 1/4 (purple down triangles) oxygen at the interface as a function of MgO thickness. The experimental results for IEC with high (green diamond) and low O_2 (orange hexagon) conditions are taken from Ref. 9.

The calculated results are summarized in Fig. 4. One can see that oxygen vacancies strongly enhance the AF IEC in agreement with previous theoretical[7] and experimental studies[9] (see Fig. 4, orange hexagons). At the same time interfacial oxidation decreases the strength of AF IEC (Fig. 4, blue up triangles) and may cause the IEC to become ferromagnetic for higher oxygen concentration (Fig. 4, purple down triangles) in partial agreement with experiment[9] (see Fig. 4, green diamonds). We included in Fig. 4 the case of the pure MTJ (black squares) in order to clearly represent the impact of oxidation on the nature of the IEC. Namely, one can note that as the amount of oxygen present in the MTJ increases, the AF IEC first decreases, vanishes and becomes finally ferromagnetic (red circles→black squares→blue up triangles→purple down triangles trend in Fig. 4).

In conclusion, we presented systematic studies of the IEC in Fe|MgO|Fe structures and showed the impact of structural relaxation and oxidation conditions on the nature of IEC in MTJs. The IEC is found to be antiferromagnetic for relaxed structures in agreement with experiment. Furthermore, it is shown that the oxygen vacancy strongly enhances the AF IEC while additional oxygen at the Fe|MgO interface, on the contrary, weakens the AF IEC and may even change its sign to FM depending on the oxygen concentration.

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