9-2006

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(Received 31 May 2006; accepted 18 July 2006; published online 12 September 2006)

Interlayer exchange coupling (IEC) in fully epitaxial Fe/MgO/Fe(001) tunnel junctions with wedge-shaped MgO layers is measured at room temperature from the unidirectional shift of the Kerr hysteresis loop. It is found that the IEC is antiferromagnetic for small MgO thickness but changes sign at 0.8 nm. *Ab initio* calculations of IEC show that this behavior can be explained by the presence of O vacancies in the MgO barrier which makes IEC antiferromagnetic for thin barriers. With increasing MgO thickness the resonance contribution to IEC from localized defect states is reduced resulting in the ferromagnetic coupling typical for perfect MgO barriers. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2349321]

Two ferromagnetic layers separated by a thin nonmagnetic spacer can be exchange coupled. The interlayer exchange coupling (IEC) was first observed for metallic spacers[1] and was found to oscillate as a function of spacer thickness.[2] Experimental observation of the IEC across an insulator has been much more challenging. There are only a few reports of measurements of IEC in magnetic tunnel junctions (MTJs).[3,4] IEC can be explained either in terms of the spin torque exerted by one ferromagnet on the other[5] or in terms of the induced density of states in the spacer by the ferromagnets.[6] For a metallic spacer the theory predicts an oscillatory coupling and relates the period of the oscillations to the spanning vectors of the Fermi surface of the spacer material. For an insulating spacer a nonoscillatory dependence is expected with the strength of the IEC exponentially decreasing as a function of barrier thickness (for a review see Ref. 7). The experimental results on MTJs (Refs. 3 and 4) significantly differ from the theoretical predictions[5,6] both in the magnitude and the sign of IEC. Changes in the electronic density of states (DOS) have crucial importance for the strength of IEC. Therefore, the presence of impurities or defects in the barrier may significantly influence IEC. It was shown that the resonant origin of the impurity-assisted IEC can make IEC antiferromagnetic.[8]

In this letter, we perform experimental and theoretical studies of the interlayer exchange coupling in Fe/MgO/Fe(001) MTJs. These junctions are interesting for applications due to very large room-temperature magnetoresistance observed experimentally.[9] Using fully epitaxial Fe/MgO/Fe(001) MTJs with wedge-shaped MgO layers we measure IEC as a function of MgO barrier thickness. In agreement with previous studies,[3] we find that IEC is antiferromagnetic for small MgO thickness but changes sign with increasing thickness. An accurate control of film roughness excludes the magnetostatic origin of this behavior. Based on *ab initio* calculations we explain this experimental fact by the presence of oxygen vacancies in MgO.

Fully epitaxial Fe/MgO/Fe(001) MTJs with wedge-shaped MgO were prepared by molecular beam epitaxy (MBE). The flatness of the interfaces was controlled by observing *in situ* the reflective high-energy electron diffraction pattern.[10] The top spin-valve sample, shown schematically in the inset of Fig. 1, was prepared as follows. After a thermal flashing of a 20×20 mm^2 single crystal MgO(001) substrate at 800 °C for 20 min, a 20 nm MgO was deposited at 350 °C to improve the surface morphology of the substrate. Then, the free bcc Fe(001) layer with thickness of 15, 20, or 30 nm was grown on the homoepitaxial MgO layer at 50 °C by a combination of a main shutter and a linear-motion shutter. Subsequently the Fe layer was annealed at 300 °C for 20 min to achieve an atomically flat surface. A single crystalline MgO(001) wedge with the thickness ranging from 0 to 2 nm was formed at 50 °C by moving the linear-motion shutter at a constant speed and annealed at 300 °C for 20 min in a vacuum of 10^{-8} Pa. During the wedge deposition, a rather low deposition rate below 0.1 Å/s was employed to prevent surface oxidation of bottom Fe layer from O and O_2 gases decomposed from evaporated MgO molecules in the MBE chamber. X-ray absorption spectroscopy and x-ray magnetic circular dichroism of the Fe/MgO inter-

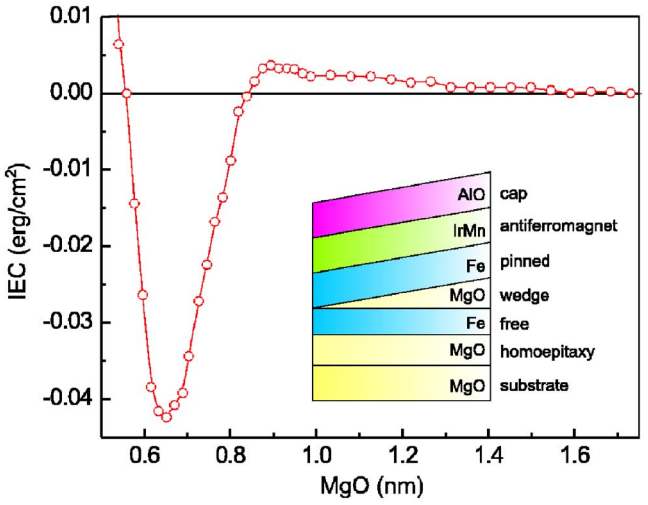

FIG. 1. (Color online) Measured IEC vs MgO thickness for the 15 nm free Fe layer. Inset shows the cross section of the sample.

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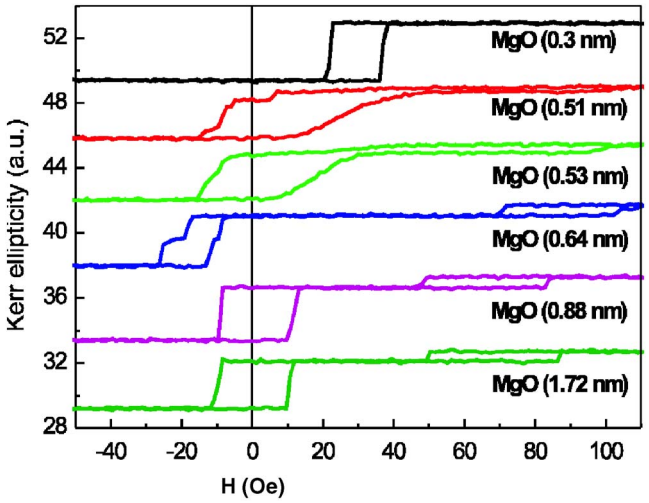

FIG. 2. (Color online) Longitudinal MOKE loops of the 15 nm free Fe layer for several MgO thicknesses.

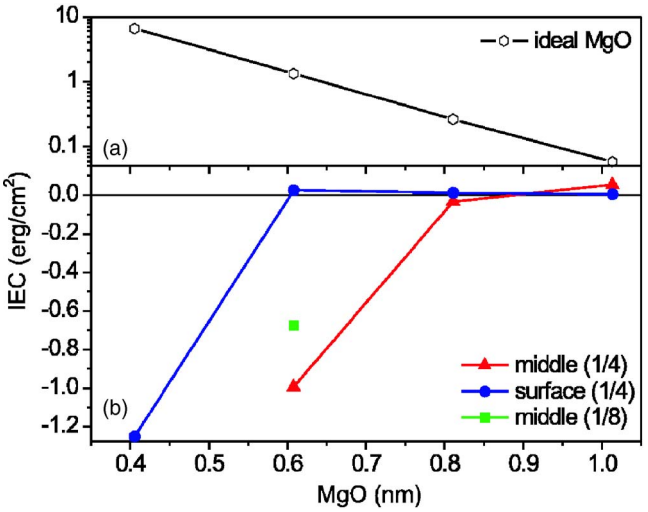

FIG. 3. (Color online) Calculated IEC vs MgO thickness for (a) ideal MgO barrier and (b) MgO with oxygen vacancies.

face show that the Fe atoms have a metallic electronic structure and a large magnetic moment which indicates that the interface is not oxidized.[11] A 10 nm Fe layer was grown at 200 °C in vacuum of 10^{-8} Pa and its magnetization was pinned by a 10 nm IrMn deposited on the Fe layer by a rf-magnetron sputtering. Finally, the whole sample was covered with a 5 nm passivated Al_2O_3 layer using the natural oxidation of Al and a reactive evaporation of Al in an O_2 atmosphere of 5×10^{-6} Torr.

Longitudinal magneto-optical Kerr effect (MOKE) was used to measure hysteresis loops of the free and pinned Fe layers at room temperature. In-plane magnetic field up to 750 Oe was applied along the [100] easy direction in the free Fe layer by a Helmholtz coil. The incident light was illuminated on the free Fe layer side through the MgO substrate. The wavelength of the incident light was 400 nm (3.1 eV) and the incidence angle was 45° with respect to normal to sample planes. The light spot (about 0.3×4 mm^2) was positioned by detecting the edges of the substrate using the reflection of the incident light. The accuracy of the light positioning was estimated to be ±0.3 mm which translates to MgO thickness accuracy of ±0.03 nm. The magnitude of IEC was obtained from the MOKE loop shift measured for various MgO thicknesses. MOKE hysteresis loops were checked against magnetization measurements by vibrating sample magnetometer for 1.8 nm of MgO.

Figure 2 shows several longitudinal MOKE loops measured for various MgO thicknesses. The loops are shifted to a positive field for 0.3 nm thickness due to strong ferromagnetic coupling of the Fe layers through pinholes. The presence of pinholes is confirmed by *in situ* UHV scanning tunneling microscopy measurements[12] as well as the temperature variation of the resistance. The maximum coupling strength is about 0.13 erg/cm^2. The coercivity increases with increasing MgO thickness up to 0.65 nm where the loops start to shift to the negative field. At the same time the loop of the pinned Fe layer shifts to the positive field. When the coupling is antiferromagnetic the MOKE loops of the two Fe layers shift in the opposite direction. Eventually, the loop of the free Fe layer shifts again to the positive field at MgO thickness of about 0.8 nm. This shows the existence of a weak ferromagnetic IEC above 0.8 nm MgO thickness. Finally, the loop becomes centered at zero field for thickness above 1.5 nm.

Figure 1 shows the dependence of IEC on MgO thickness. The coupling is large ferromagnetic for MgO thickness below 0.3 nm due to pinholes. It becomes antiferromagnetic for MgO thickness range of about 0.5–0.8 nm; then it reverses to weak ferromagnetic above 0.8 nm. Finally, the IEC gradually approaches zero. We find that the absolute value of IEC increases with increasing Fe free layer thickness (15, 20, 30 nm). This indicates that the better quality and better interface flatness of the MgO wedge, achieved by increasing the thickness of the bottom Fe layer, may provide a stronger antiferromagnetic coupling. However, the values of the coupling observed in Ref. 3 are still much larger than ours which may be attributed to larger concentration of oxygen vacancies.

It should be pointed out that in Ref. 3 the coupling for larger barrier thickness approaches a finite value. This is attributed to the "orange peel" effect which given the quality of the sample is nearly constant at this thickness range. In our case the coupling approaches zero in the same range of thicknesses (above 1.5 nm). Therefore the magnetostatic interaction due to interface roughness is negligible as a consequence of the high-quality Fe/MgO interfaces and the small area of the sample at a given thickness. This excludes the orange peel coupling effect as the origin of the sign change of IEC.

In order to explain the observed variation of the IEC as a function of MgO thickness, we have performed *ab initio* calculations of the coupling for ideal Fe/MgO/Fe MTJs and for junctions which contain O vacancies. This is motivated by the fact that O vacancies are a common type of defects in MgO-based MTJs.[13] On the other hand, as follows from the theoretical predictions, localized defect states in the barrier can mediate antiferromagnetic IEC.[8]

IEC is a difference in the total energy between antiparallel and parallel magnetizations of a MTJ, $J=(E_{AP}-E_P)/2$. We use a pseudopotential plane-wave method[14] to calculate the total energy. For an ideal MgO barrier we find that IEC is ferromagnetic and decays almost perfectly exponentially with MgO thickness, as is seen from Fig. 3(a). From the slope of the graph we find that the decay rate of IEC is

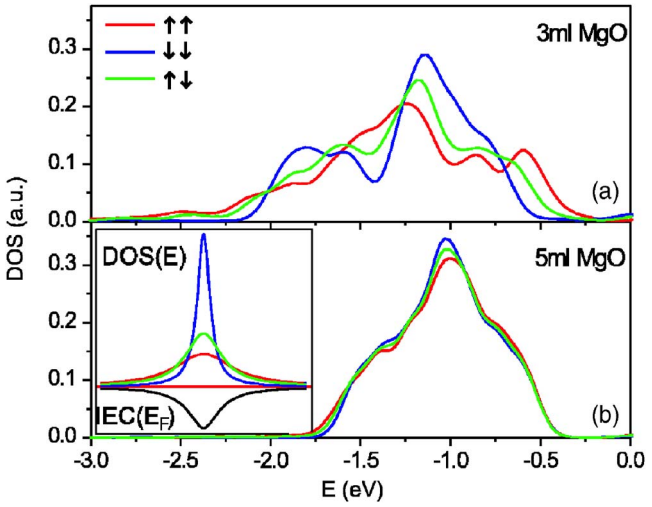

FIG. 4. (Color online) DOS on the vacancy site for (a) 3 ML and (b) 5 ML of MgO. The Fermi energy (E_F) lies at zero. Inset shows the model vacancy DOS and calculated IEC as a function of E_F.

0.4 Å^{-1}. This value agrees very well with the slope of the conductance against MgO thickness[15] and coincides with the decay rate of the Δ_1 state in the complex band structure of MgO.[16] This indicates that the main contribution to IEC comes from the electrons around the Fermi surface. This behavior is in agreement with theoretical predictions[6] but at odds with our experimental data.

To include O vacancies in the calculation we consider a supercell with one vacancy per four or eight oxygen atoms in the plane. Applying this approach to bulk MgO produces an *F* center (state with two electrons trapped in the cavity formed by removing a neutral O atom)[17] close to the middle of the band gap. The width of the level depends strongly on the vacancy density and is about 1 eV for the $\frac{1}{4}$ concentration. We calculate IEC assuming that O vacancies lie within 1 ML of MgO, either in the middle or on the surface of the MgO slab. Figure 3(b) shows the calculated IEC versus MgO thickness in the presence of O vacancies. It is seen that the coupling is antiferromagnetic for small MgO thickness but changes sign at larger thickness. IEC is stronger when the vacancy lies in the middle and increases with vacancy concentration. At large thickness the coupling becomes comparable to that of the ideal system. This behavior is consistent with our experimental observations.

The fact that IEC changes from ferromagnetic to antiferromagnetic in the presence of vacancies indicates that the antiferromagnetic IEC is mediated by the resonance coupling through the *F* center.[8] To provide a simple qualitative picture of this phenomenon we estimate IEC by calculating the change in the DOS due to coupling of the vacancy level to the ferromagnets. The vacancy DOS is approximated by a Lorentzian $D_\gamma(\epsilon)=(\gamma/2\pi)/[(\epsilon-\epsilon_0)^2+(\gamma/2)^2]$, where ϵ_0 is the center of the spectrum and γ is its width. The level width is composed of an intrinsic part γ_0 (due to coupling to the other vacancies) and a spin dependent part γ_σ (due to coupling to the ferromagnets). From Fig. 4, where DOS on the vacancy level is shown, it is clear that for 5 ML of MgO the width of the vacancy state is γ_0 because it does not depend on the magnetization orientation [Fig. 4(b)]. This is due to the relatively large MgO thickness which makes the coupling between the ferromagnets and the vacancy weak. In that case the vacancy does not affect IEC which is ferromagnetic and very close in value to the ideal case. On the other hand for 3 ML of MgO [Fig. 4(a)] the coupling to the ferromagnets is strong and the vacancy level width becomes spin dependent, $\gamma=\gamma_0+\gamma_\sigma$. Note that $\gamma_{\uparrow\uparrow}>\gamma_{\uparrow\downarrow}>\gamma_{\downarrow\downarrow}$ due to the fact that the escape rate for majority- and minority-spin electrons is controlled by tunneling through the Δ_1 and Δ_5 complex bands of MgO, respectively, which have very different decay rates.[16,18] Thus, the changes of energy due to coupling to the ferromagnets are[6] $\Delta E=-\int_{-\infty}^{\epsilon_F}d\epsilon(\epsilon-\epsilon_F)[D_\gamma(\epsilon)-D_{\gamma_0}(\epsilon)]$ and $\mathrm{IEC}=-\int_{-\infty}^{\epsilon_F}d\epsilon(\epsilon-\epsilon_F)[D_{\gamma_{\uparrow\uparrow}}(\epsilon)+D_{\gamma_{\downarrow\downarrow}}(\epsilon)-2D_{\gamma_{\uparrow\downarrow}}(\epsilon)]$. One can see that when the level is not coupled to the ferromagnets ($\gamma=\gamma_0$) the resonant contribution to IEC is identically zero. However, when the levels are broadened the asymmetry allows for negative values of the coupling. This is evident from the inset of Fig. 4 which shows the result of the model calculation, indicating antiferromagnetic IEC when the defect state is partially occupied.

In conclusion, we have observed interlayer exchange coupling in fully epitaxial Fe/MgO/Fe(001) MTJs. The IEC changes sign from antiferromagnetic to ferromagnetic with MgO thickness. *Ab initio* calculations explain the antiferromagnetic coupling and the change of the IEC sign by the presence of oxygen vacancies in MgO.

The research at AIST was partly supported by Nanotech Challenge Project of the New Energy and Industrial Technology Development Organization (NEDO). The research at UNL was supported by NSF (Grants Nos. MRSEC DMR-0213808 and DMR-0203359) and the Nebraska Research Initiative.

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