Inverse and oscillatory magnetoresistance in Fe(001)/MgO/Cr/Fe magnetic tunnel junctions

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The effect of Cr(001) insertion layers in Fe(001)/MgO/Cr/Fe magnetic tunneling junctions is studied from first principles. It is shown that with the increase in the Cr(001) layer thickness, the tunneling magnetoresistance (TMR) first decreases rapidly and then oscillates with a two-monolayer period. At some thicknesses, the oscillation leads to a sign reversal of the TMR. The oscillatory interfacial Cr moment at the Cr-MgO interface as a function of the Cr layer thickness, which arises from the layer-antiferromagnetic ordering of Cr, is the cause for the oscillatory TMR.

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Interlayers in magnetic tunnel junctions (MTJs) are layers with atomic thicknesses inserted between one of the magnetic electrodes and the barrier. Such interlayers can have a significant effect on spin-dependent tunneling. In addition to the obvious possibility that they can be used to further optimize the performance of the MTJs, other effects can also arise. First, the interlayer acts as the additional scattering layers and its band structure plays an important part in the tunneling process. Second, the interface properties between the magnetic electrodes and the barrier, such as the interface resonance states, are modified by the interlayers. For example, quantum well states may form within the interlayers, leading to an oscillatory tunneling magnetoresistance (TMR) as shown in an earlier experiment[1] using a Cu interlayer in amorphous Al-O tunnel junctions. For MgO-based MTJs, the interlayer effects may be amplified because of the coherent tunneling process. The first-principles calculation[2,3] shows that with ultrathin Ag and Co interlayers at the interface in Fe/MgO/Fe MTJs, the TMR is enhanced due to the suppression of the interface resonance states. Other more recent theoretical works have also considered Ag (Ref. 4) and V (Ref. 5) as interlayer materials in Fe/MgO/Fe-based MTJs.

As a spacer layer, the Cr(001) presents some unique properties because of the absence of the Δ_1 band at the Fermi energy. This is expected to impact significantly the TMR in MgO-based junctions because the fully spin-polarized Δ_1 band at the Fermi energy in Fe or FeCo is the primary reason for producing the large TMR.[6] Furthermore, the Cr(001) on Fe(001) has the layer-antiferromagnetic (LAF) ordering.[7] Scattering caused by such magnetic ordering near the interface may also have a profound impact on the TMR. Observed effects range from an oscillatory TMR with two-monolayer (ML) period as a function of the Cr thickness in FeCo/Al-O/Cr/Fe junctions,[8] to a monotonous decrease in the TMR as a function of the Cr thickness in single-crystal Fe(001)/MgO/Cr/Fe MTJs.[9] A more recent study[10] in Fe/MgO/Cr/Fe junctions also observed a two-monolayer period in the TMR oscillation, but with an opposite oscillation phase than the result from the Al-O-based junctions.[8] These seemingly conflicting experimental results call for a careful study from the first-principles calculations.

In this paper, we present a first-principles study of the Cr(001) interlayer effect in Fe(001)/MgO/Cr(001)(0–11ML)/Fe MTJs. The calculated tunneling conductance and TMR show a two-monolayer oscillation. The origin of such oscillation is tracked to the oscillatory moment orientation in the Cr layer. We also show that the phase of such oscillation depends critically on the interface properties at the Fe/Cr interface. In particular intermixing of Fe and Cr atoms at the interface layer can lead to a reversal of the moments in the Cr layers, which, in turn, changes the phase of the TMR oscillation. Thus our calculation will be able to offer a consistent explanation for all experimental observations.

In our calculation, the MgO barrier is fixed at 12 ML, or 2.3 nm. This thickness is significantly thicker than that used in past calculations.[6,11,12] As pointed out by Butler *et al.*,[6] thicker MgO barrier MTJs can significantly suppress the relative conductance contribution from interface resonance states. We assume that there is no vertical relaxation and that all the layers stack epitaxially. The Cr interlayer is varied from 0 to 11 ML. There is very little lattice mismatch between the Fe and Cr, so we use the lattice constant of the Fe substrate (2.866 Å) for the Cr layer. The in-plane lattice constant of MgO is $\sqrt{2}$ larger than that of the Fe while the interlayer MgO spacing is 2.21 Å, a value taken from experiment.[13] Both the Fe-O distance and Cr-O distance are assumed to be 2.163 Å. In order to fill the space with atomic spheres, empty spheres with a radius of 0.877 Å are inserted on both interfaces of the MgO barrier. The self-consistent electronic structure and tunneling conductance calculations are performed using the layer Korringa-Kohn-Rostoker method,[14] based on the local-spin-density approximation[15] and the Landauer-Büttiker formula,[16,17] in the same manner as in the previous calculations.[6,11]

The magnetic structure of a Fe(001)/MgO/Cr/Fe MTJ with 4 ML Cr is illustrated in Fig. 1(a). The red and blue arrows label the moment directions for the parallel (P, when the magnetic moments of the Fe electrodes are aligned parallel to each other) and antiparallel (AP, when the magnetic moments of the Fe electrodes are aligned antiparallel to each other) configurations. Figure 1(b) shows the calculated layer-resolved magnetic moment for the same structure. The moments on the MgO layers are negligible. For the P configu-

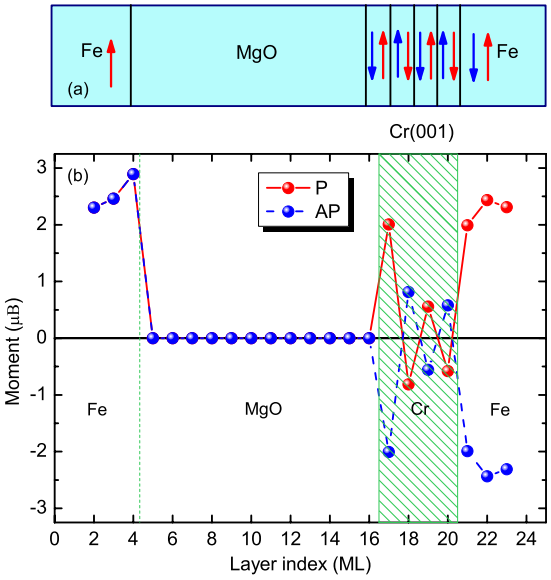

FIG. 1. (Color online) (a) Schematic sketch of the structure of a Fe/MgO/Cr/Fe tunneling junction. The arrows indicate the magnetic moment directions. (b) The layer-resolved magnetic moment for parallel alignment (red solid line) and antiparallel alignment (blue dashed line) of the Fe electrodes for a junction with 4 ML Cr (green shadow region).

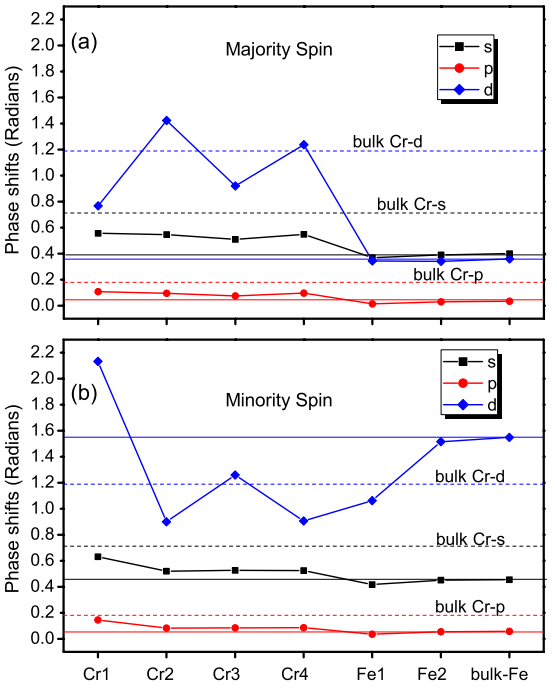

FIG. 2. (Color online) The *s*, *p*, and *d* phase shifts for each Cr and Fe layer in Fe/MgO/Cr4/Fe MTJ in the (a) majority and (b) minority spin channels. The Cr1–4 are the four Cr layers and Cr1 is the interfacial Cr layer next to MgO. The Fe1–3 are the three Fe layers adjacent to Cr. The horizontal dashed lines indicate the corresponding values for bulk Cr and the solid lines are those of bulk Fe.

ration, starting from the Fe-Cr interface the moments of the Cr layers are −0.58 μ_B, 0.56 μ_B, −0.82 μ_B, and 2.01 μ_B, respectively. Independent of the thickness of the Cr layer (0–11 ML), the Cr moment at the Fe-Cr interface is always antiferromagnetically coupled to the adjacent Fe electrode, and the moments of the rest of the Cr layers alternate in sign layer by layer. The calculated LAF magnetic ordering of Cr layers in Fe/MgO/Cr/Fe MTJs is similar to the observed magnetism in Cr thin films on Fe(001) substrate.[7]

The interfacial moment of Cr at the Cr-MgO interface with increasing Cr thickness stays approximately constant and has a well-defined magnetic moment (2 μ_B). This value is far greater than the moment of bulk Cr and it is almost as large as the moment of bulk Fe. This is an important feature that will impact the transport properties. The large moment reflects the shift of the Cr *d* bands relative to the Fermi energy. The bulk bcc Cr and Fe have nearly the same bands that are filled with different numbers of electrons. The paramagnetic Cr has 12 electrons in each spin channel, compared to 14 electrons in Fe majority channel and 12 electrons in the Fe minority channel. The position of the Fermi energy in Cr is such that its band structure matches very well with the Fe minority channel but not with the Fe majority channel. As we already noted, there is no majority spin Δ_1 band at the Fermi energy in Cr(100). Naively, this would lead to a significant reduction in the TMR. However, because of its magnetic moment of 2 μ_B, the interfacial Cr layer has 13 majority electrons and 11 minority electrons. This brings the number of electrons in the majority spin channel a lot closer to bulk Fe while removing the matching of the bands in the minority spin channel. Its effect on transport can be predicted[18] from the *s*, *p*, and *d* phase shifts of the self-consistent potential plotted in Fig. 2. From the plot we see that for the majority spin channel all phase shifts for the interfacial Cr layer (next to the MgO) are about halfway between the corresponding phase shifts of bulk Fe and bulk Cr whereas in the minority spin channel the difference between the interfacial Cr layer phase shifts and the bulk Fe phase shifts are essentially the same as those between bulk Cr and bulk Fe. As a consequence, the interfacial Cr layer acts more like an Fe layer, and the tunneling conductance depends strongly on the spin orientation of this layer relative to the Fe electrodes, as we will see below.

Using the converged electronic structure, the tunneling conductances are calculated using 1024×1024 $\mathbf{k}_\parallel$ points in the two-dimensional Brillouin zone (2DBZ). The calculated TMR ratio (defined as $G_P/G_{AP}-1$) and the tunneling conductance are shown in Fig. 3. The TMR ratio first decreases rapidly with the increasing Cr thickness. After the Cr layer thickness reaches 4 ML, the TMR ratio shows an oscillation. For the parallel configuration, the conductance decreases exponentially with the Cr thickness up to 4 ML, then oscillates with a two-monolayer period with peaks at even layers and valleys at odd layers. Similarly, for the AP configuration, the conductance also oscillates with two-monolayer period, but its phase is opposite of that of the P configuration. If the Cr layer thickness is more than 4 ML, the TMR oscillates between positive and negative values.

The origin of the decay and the oscillation in the conductance is tracked to the dependence of the transmission probability on the Cr layer thickness as shown in Figs. 4 and 5. Hereafter we define "majority" and "minority" in both con-

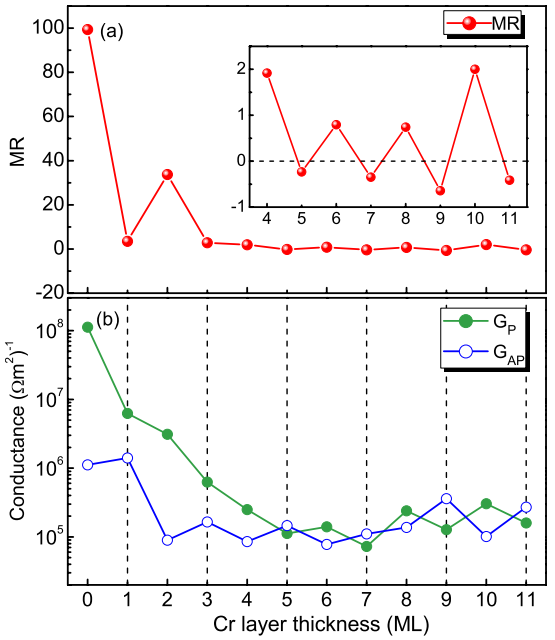

FIG. 3. (Color online) (a) MR as a function of Cr layer thickness (the inset shows the details for the range of 4–11 ML). (b) Tunneling conductance in P (circle) and AP (open circle) configurations.

figurations as the electron spin parallel and antiparallel to the magnetic moment of the left Fe electrode as shown in Fig. 1(a). First, we examine the contribution from the Fe(001) Δ_1 band. The majority Δ_1 electron has the slowest decay rate in the MgO barrier and it makes up the main contribution to the conductance of the P configuration.[6] However, the Δ_1 band in Cr is far above the Fermi energy thus the Cr layer acts as a barrier for the majority Δ_1 electrons in the P configuration,[8,12] causing a rapid decay of the transmission probability around the $\bar{\Gamma}$ point in 2DBZ. This is clearly shown in Fig. 4, (1a)–(1f). 4–5 ML Cr is sufficient to totally suppress the majority Δ_1 electron tunneling. This explains the decrease in the conductance in Fig. 3(b) for the P configuration and the decrease in the TMR.

The conductance oscillation for the P configuration arises mainly from the minority spin channel, and the oscillation for the AP configuration arises mainly from the majority spin channel. In the following discussion we will focus on these two channels. Without the Cr interlayer, the parallel minority conductance is mainly from the Bloch states around the $\bar{X}$ point at the boundary of the 2DBZ and the antiparallel majority conductance is mainly from the Bloch states around the $\bar{\Gamma}$ point.[6] As shown in Figs. 4 and 5, when the Cr layer is inserted, these Bloch states still dominate the transmission for the P minority and AP majority channels. The two-monolayer oscillation with the increasing of the Cr layer thickness is clearly seen in the transmission probabilities in the 2DBZ but the largest oscillations are seen near the two special k points mentioned above.

To clarify the origin of this transmission and conductance oscillation, we examine the so-called tunneling density of states (TDOS). This is defined as the density of electronic states that are matched to an incident Bloch state with unit flux. It reflects how the Bloch state are transmitted through each layer.[6] First, we look at the AP majority channel at $\mathbf{k}_{\parallel} = (0.025, 0.025)$, a point inside the region in 2DBZ that exhibits large oscillations, as shown in Fig. 5, (1a)–(1f). The thickness dependence of layer-resolved TDOS at this $\mathbf{k}_{\parallel}$ point is plotted in Fig. 6(a) for 5–8 Cr interlayers. The magnitude of TDOS in the interfacial Cr layer also shows large oscillation with respect to the Cr layer thickness as shown in the inset and is strongly correlated with the final transmission probability.

The correlation between the interfacial Cr moment direction and the tunneling current is further confirmed when we flip the interfacial Cr moment artificially while keeping the moment on other Cr layers the same. We flip the moment of the interfacial Cr layer for the cases of 5–8 ML Cr insertion layers and calculate the minority conductance in the P configuration. The results are shown in Fig. 6(b) which shows that the oscillation of the conductance is completely reversed as well. This suggests that the interfacial Cr layer indeed acts like an Fe layer, and when its moment is aligned parallel to the moment of the Fe electrode on the other side of the

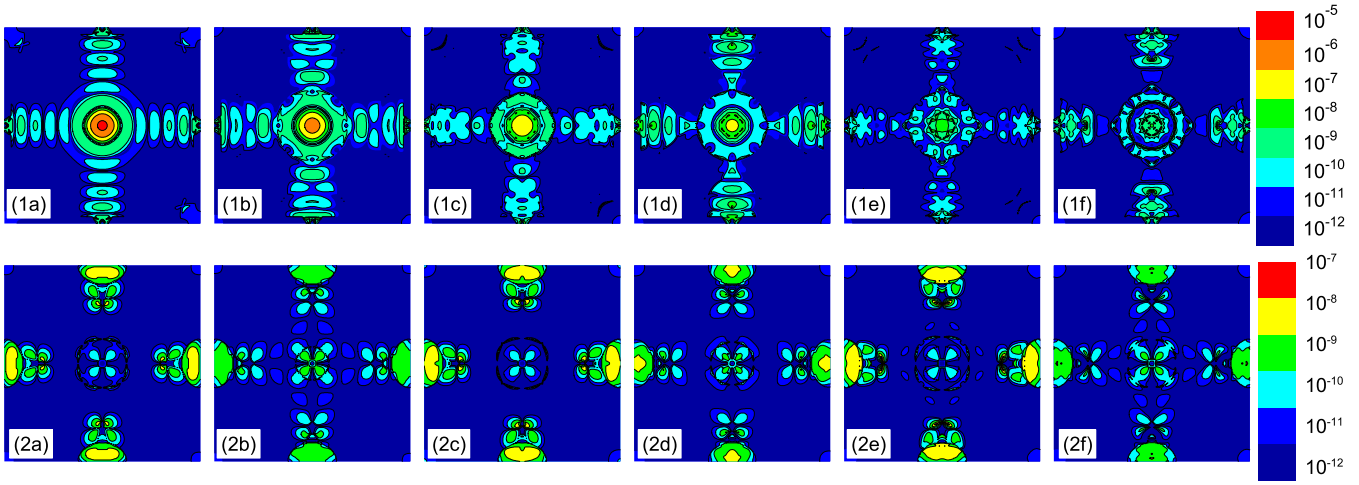

FIG. 4. (Color online) Transmission probability in 2DBZ with 256×256 k mesh for Cr from 2 to 7 ML for P configuration (left to right), (1a)–(1f) for the majority spin (upper color scale), (2a)–(2f) for the minority spin (lower color scale).

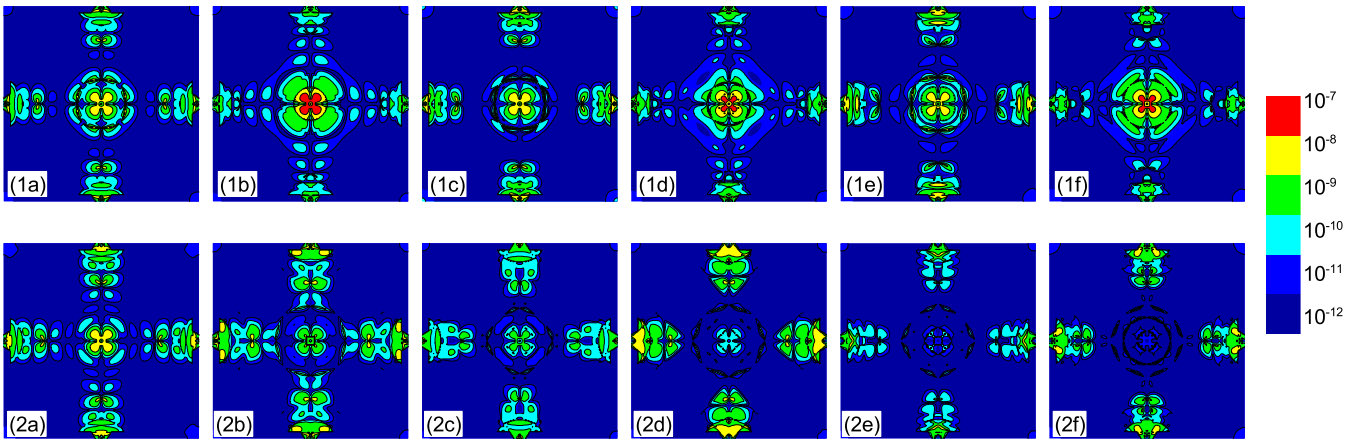

FIG. 5. (Color online) Transmission probability in 2DBZ with 256×256 k mesh for Cr from 2 to 7 ML for AP configuration (left to right), (1a)–(1f) for majority electrons in the left Fe electrode, (2a)–(2f) for minority electrons in the left Fe electrode.

barrier, the tunneling conductance is increased. From this we conclude that the oscillation of the conductance come from the oscillatory magnetic alignment of the interfacial Cr layer.

To further prove the significance of the moment of the interfacial Cr layer, we calculate the tunneling conductance with 1 ML Mg inserted at the MgO-Cr interface. The addition of the Mg layer greatly reduces the interfacial Cr moment, as shown in the inset of Fig. 7. Now the number for the interfacial Cr layer is closer to that of bulk Cr rather than bulk Fe. If the oscillation in the conductance is connected to the oscillatory Cr moment, then in this case the oscillation should diminish since the Cr moment is almost zero. This is indeed what we see in Fig. 7, which shows that the oscillation of the P minority conductance is decreased by nearly an order of magnitude with the addition of the Mg monolayer.

The decrease and the two-monolayer period oscillation of the calculated TMR ratio agree well with recent experiments.[8–10] However, compared to the most relevant experiment with the MgO barrier,[10] the calculated TMR oscillation appears to be out of phase. The theory predicts TMR peaks at even layers and troughs at odd layers, but the experiment showed the opposite. It was suggested that CrO formation at the Cr/MgO interface[19] is the most likely defect structure. This might account for a shift of the oscillation period. The LDA used in our calculation does not properly treat the strongly correlated nature of CrO electronic structure thus this question will have to be answered in a future study. On the other hand, a similar phase shift of interlayer exchange coupling in Fe/Cr/Fe was attributed to the possible intermixing of Fe-Cr.[20,21] A complete reversal of the Cr moments may occur with a small amount of intermixing of Cr and Fe at the Fe/Cr interface.[20,21] Although in the experiment of Fe/MgO/Cr/Fe (Ref. 10) such intermixing was deemed unlikely, small amount of intermixing within a monolayer is still plausible. We carried out a coherent potential approximation[22] total-energy calculation for the MTJ structure in which a two monolayer Fe/Cr interface is intermixed, in the manner $Fe/MgO/Cr_3/Cr_xFe_{1-x}/Fe_xCr_{1-x}/Fe$, where x is the concentration of intermixing. Figure 8(a) shows the change in the total energy due to intermixing. In this figure the label LAF denotes the same magnetic configuration in the Cr layers as the unmixed case (shown in Fig. 1) and reversed LAF denotes the configuration in which all magnetic moments are reversed in Cr layers. Figure 8(a) shows the energy difference between the LAF and reversed LAF magnetic configurations in Cr layers. Compared to the LAF configuration, when the intermixing exceeds 30%, the reversed LAF configuration becomes energetically favorable.

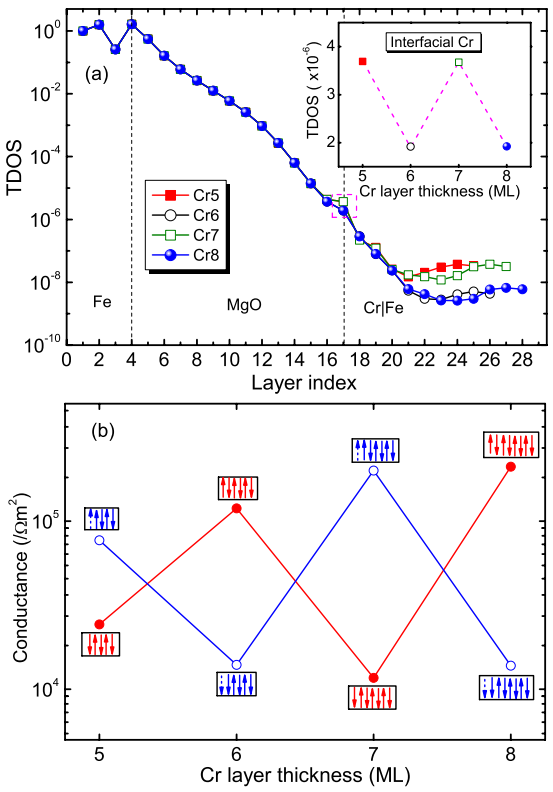

FIG. 6. (Color online) (a) TDOS for majority in AP configuration for Fe/MgO/Cr(5–8 ML)/Fe at $\mathbf{k}_{\parallel}=(0.025,0.025)$. The pink dashed square labels the interfacial Cr. The inset shows the interfacial TDOS at this $\mathbf{k}_{\parallel}$ point. (b) The minority conductance in P configuration with (open circle) and without (solid circle) filliping the interfacial Cr moment for Fe/MgO/Cr(5–8 ML)/Fe. The arrows denote the moment direction of the Cr at the Cr-MgO interface.

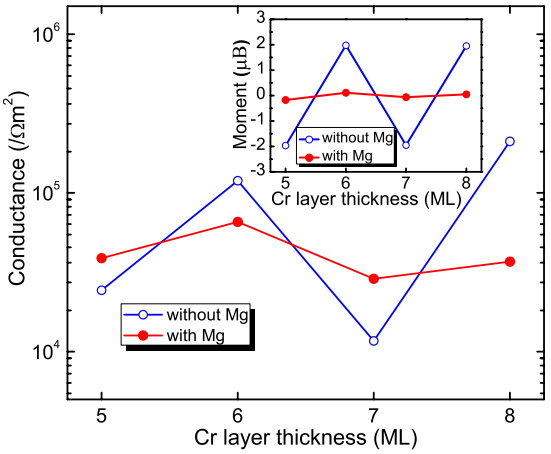

FIG. 7. (Color online) The minority conductance in P configuration with (solid circle) and without (open circle) a monolayer Mg interlayer in Fe/MgO/Cr(5–8 ML)/Fe. Inset shows the interfacial Cr moment.

Such a change in the magnetic-moment orientations is similar to the Cr overlayers on Fe(001) substrate,[20,21] where the transition occurs at a concentration between 25% and 33% for 3 ML Cr on Fe. We plot the ground-state magnetic moment in the first 4 ML of Cr as a function of the interface mixing in Fig. 8(b). The interfacial moment of Cr is flipped when the mixing exceeds 30%. Based on the above discussion, the conductance and TMR oscillation phase should also flip.

In conclusion, through first-principles calculations of the electronic, magnetic, and tunneling properties of Fe/MgO/Cr/Fe MTJs, we show that the tunneling conductance is strongly dependent on the Cr moment at the Cr-MgO interface. The calculated conductance as well as the MR ratio show two-monolayer oscillations as a function of Cr layer thickness, in agreement with experiments. We identify that the oscillatory interface moment of Cr due to the LAF magnetic ordering is the origin of this oscillatory TMR effect.

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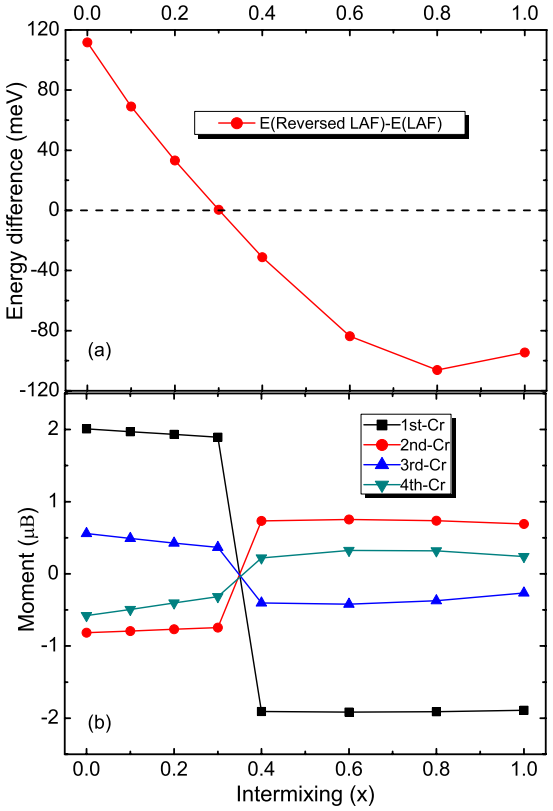

FIG. 8. (Color online) (a) The energy difference between the LAF and reversed LAF magnetic ordering in Cr layers of $Fe/MgO/Cr_3/Cr_xFe_{1-x}/Fe_xCr_{1-x}/Fe$. (b) The ground-state magnetic ordering in the first four monolayers of Cr in $Fe/MgO/Cr_3/Cr_xFe_{1-x}/Fe_xCr_{1-x}/Fe$.

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