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Tailoring TMR ratios by ultrathin magnetic interlayers: A first-principles investigation of Fe/MgO/Fe

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ABSTRACT

For spintronic device applications, large and in particular tunable tunnel magnetoresistance (TMR) ratios are inevitable. Fully crystalline and epitaxially grown Fe/MgO/Fe magnetic tunnel junctions (MTJs) are well suited for this purpose and, thus, are being intensively studied [1]. However, due to imperfect interfaces it is difficult to obtain sufficiently large TMR ratios that fulfill industrial demands (e.g. [2]).

A new means to increase TMR ratios is the insertion of ultra-thin metallic buffer layers at one or at both of the Fe/MgO interfaces. With regard to their magnetic and electronic properties as well as their small lattice mismatch to Fe(001), Co and Cr spacer are being preferably investigated.

We report on a systematic first-principles study of the effect of Co and Cr buffers (with thicknesses up to 6 ML) in Fe/MgO/Fe magnetic tunnel junctions (MTJs) on the spin-dependent conductance. The results of the transport calculations reveal options to specifically tune the TMR ratio. Symmetric junctions, i.e. with Co buffers at both interfaces, exhibit for some thicknesses much larger TMR ratios in comparison to those obtained for Fe-only electrodes. Further, antiferromagnetic Cr films at a single interface introduce TMR oscillations with a period of 2 ML, a feature which provides another degree of freedom in device applications. The comparison of our results with experimental findings shows agreement and highlights the importance of interfaces for the TMR effect.

INTRODUCTION

Fully crystalline Fe/MgO/Fe MTJs show very high TMR ratios [3-6]. After intensive studies of these systems, the research was gradually extended to other promising systems. MgO tunnel junctions with amorphous CoFeB electrodes for instance were found to improve structural and magnetic properties, resulting in giant TMR ratios [1].

The detailed structure of the interfaces in Fe/MgO/Fe essentially determines the spin-polarized current. Thus, it is obvious to manipulate the interfaces in a controlled way to achieve larger TMR ratios. Considering the magnetic profiles in Figs. 1 and 2, it is expected that Cr and Co buffers have a sizable effect on the tunnel current, and especially on its spin-polarization.

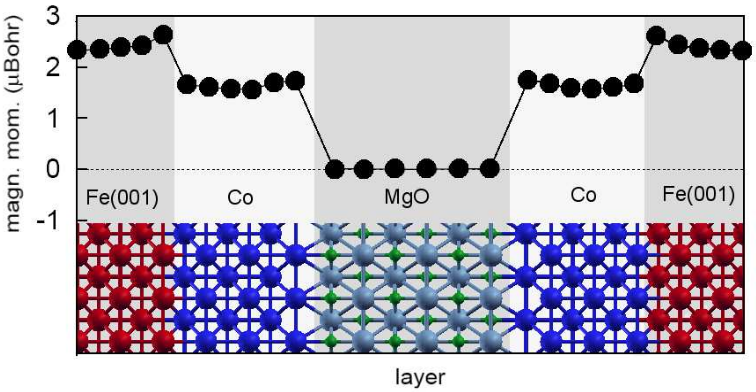

Figure 1. Layer-resolved magnetic moments of bcc Fe(001)/*x*(Co)/6MgO/*x*(Co)/Fe(001) magnetic tunnel junction with *x* = 6. The geometry of the MTJ is sketched at the bottom.

Magnetic tunnel junctions with bcc Co electrodes are theoretically predicted [7] to provide much larger TMR ratios than those with Fe electrodes. However, Co grows only up to a few monolayers on MgO in the bcc phase; for thicker layers a structural transition to the hcp structure takes place, thus introducing imperfections which definitely reduce the TMR ratio. One aim of this work is to investigate whether thin Co interlayer in Fe/MgO/Fe increase the TMR ratios, similar to those ratios predicted with infinite Co leads and comparable with those obtained with CoFeB electrodes.

Nagahama et al. [8] showed that the insertion of Cr films in MTJs with amorphous AlO tunnel barriers and Fe leads exhibits a 2-ML oscillation of the experimental TMR ratio as a function of Cr thickness. A 2-ML oscillation is a signature of a layerwise antiferromagnetic order in the Cr film, in agreement with theoretical findings for Mn buffers [9]. In the latter work, the even-odd effect in the sign of the TMR ratio was attributed to the atomic Mn layer adjacent to the tunnel barrier: its magnetization direction plays a key role in the spin-dependent electronic transport.

Cr couples antiferromagnetically to Fe(001) and shows layerwise AFM order (Fig. 2). In addition, a large magnetic moment is found at the interface with MgO. This finding raises the question whether a single Cr spacer at a single interface produces a defined sign reversal of the TMR ratio.

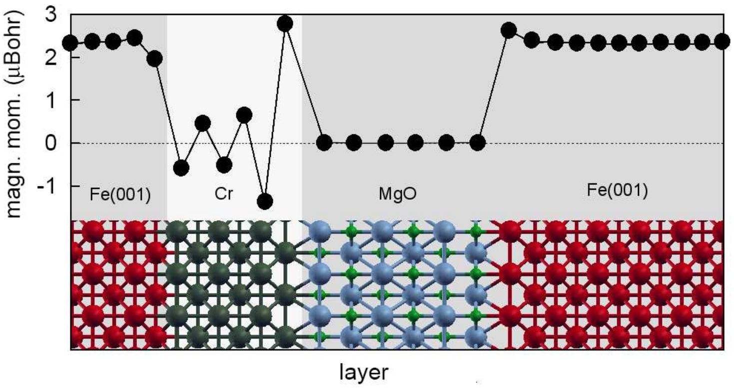

Figure 2. As Figure 1, but for bcc Fe(001)/*x*(Cr)/6MgO/ Fe(001) MTJ with *x* = 6.

THEORETICAL BACKGROUND

In a first step, *ab-initio* electronic-structure calculations were performed within the framework of the local spin-density approximation to density functional theory. The so achieved self-consistent potentials serve as input for the subsequent transport calculations. Both electronic-structure and transport properties are obtained by a Korringa-Kohn-Rostoker multiple-scattering Green's function formalism. Due to its perfect adaptation to the planar geometry a layer-KKR computer code was used for the electronic transport calculations. The self-consistent treatment of the Fe(001)/x(Co)/MgO/x(Co) /Fe(001) and Fe(001)/ x(Cr)/MgO/Fe(001) MTJs, x = 1, …, 7 ML, follows closely those for Fe(001)/MgO/Fe(001) reported in [10]. In particular, atomic positions and interlayer distances were taken from experiment [11]. So, slight changes are expected due to the different atomic volumes of Co and Cr with respect to Fe. The number of MgO layers was fixed for each set-up to 6 ML (corresponding to a thickness of 10.7 Å).

Within the Landauer-Büttiker approach [12], the zero-bias conductance is calculated in terms of the transmittances $T(E)$ at the Fermi level. The latter is computed by integrating the wavevector-resolved transmittances $T(E, k_{||})$ over the two-dimensional Brillouin zone (2BZ) [13], where $T(\mathrm{E}, k_{||})$ is the sum of the transmission probabilities of all Bloch states in the leads. Since both setups exhibit *4mm* symmetry, the number of wavevectors $k_{||}$ in the 2BZ integration was reduced from 80 000 equidistant mesh points to about 10000 of the irreducible part while maintaining the same level of accuracy.

The TMR ratio is expressed by the asymmetry of the conductances for the parallel (G_P) and antiparallel (G_{AP}) magnetic configurations of the Fe electrodes, normalized by the conductance of the AP case (`optimistic TMR ratio').

RESULTS AND DISCUSSION

Co interlayers at both Fe/MgO interfaces

The conductance for the parallel configuration G_P is almost constant with an apparent 2-ML oscillation, with maximum (minimum) conductance for an even (odd) number of x ML. In contrast G_{AP} shows a more complex thickness dependence (Fig. 3). G_{AP} starts approximately two orders of magnitudes smaller than GP at x = 0 ML, but reaches a pronounced maximum an order of magnitude larger at a thickness of two Co layers. For x = 3 – 5 ML it decreases and reaches nearly the level obtained without Co spacers. Another, but some smaller maximum is obtained for one additional Co layer (x = 6 ML). In comparison to the Fe/MgO/Fe MTJ without Co film, a sizably smaller G_P but larger G_{AP} value is achieved for infinite Co electrodes.

The calculated TMR ratios exhibit three noticeable characteristics. Firstly, 3 and 5 ML thick Co interlayers lead to huge TMR ratios: 10000% at 3 ML and 15700% at 5 ML – which are significantly larger than the 6800% obtained without Co spacers (indicated by the green horizontal line in Fig. 3). Secondly, the TMR at 2 ML Co drops as a consequence of the large G_{AP} value, caused by interface resonances, nearly to zero. Thirdly, a much smaller TMR value is calculated for infinite Co leads in comparison to that determined with pure Fe leads, a finding in contrast to results reported in [7]. This may be related to differences in the geometries. Previous investigations of Fe/MgO/Fe systems have shown that slightly differing atomic positions in the interface region can lead to sizably different conductances and TMR ratios.

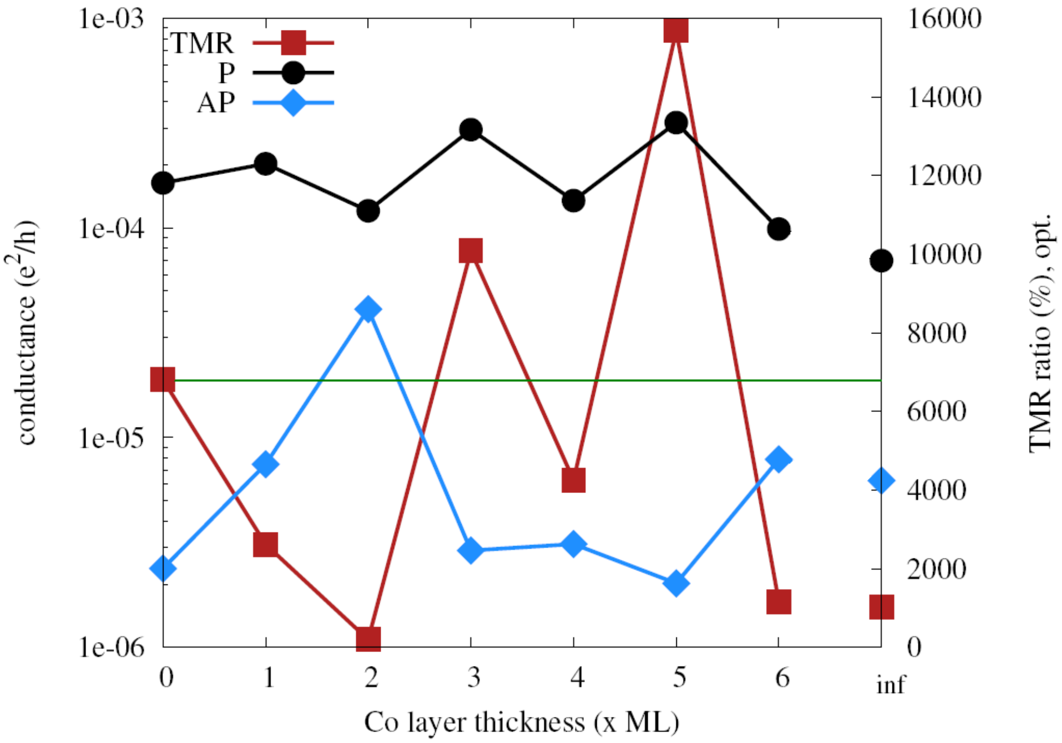

Figure 3. Conductances for the P (black circles) and AP (blue diamonds) magnetic configurations of Fe/*x*(Co)/MgO/*x*(Co)/Fe versus Co thickness. The `optimistic' TMR ratio is shown as red squares. Results for Co electrodes, replacing the Fe electrodes are shown in addition (`inf').

Cr interlayers at one Fe/MgO interface

Fig 4a. displays in analog to Fig 3., the thickness dependence of the P and AP conductance with up to 7 Cr layers. Both, G_P and G_{AP} exhibit an exponential decay as a function of the Cr thickness *x*. The G_P decay rate is hereby visibly larger than that one for G_{AP}.

G_P and G_{AP} reveal superimposed to the exponential decay, even-odd oscillations that are in antiphase. These characteristics can be traced back to the layer-wise antiferromagnetism of the Cr layers. In Fig 2. the exemplary case with $x = 6$ ML shows that Cr couples layer-wise antiferromagnetically to the Fe(001) substrate. It turns out and can be seen too in Fig 2. that the magnetic layer at the Cr/MgO interface possesses the largest uncompensated local magnetic moment of the Cr spacer. It can be deduced from previous tunnel magneto-resistance investigations with layer-wise antiferromagnetic Mn films [9] that the Cr film acts generally as a spin-filter for the electron currents. But the decisive influence which changes the spin-polarization of the currents can be directly addressed to the magnetic atom adjacent to the MgO barrier.

With help of Fig 4b, which displays the local magnetizations of these interface layers, it is clear that an even number of Cr layers leads to a positive local magnetic moment which results directly in local maxima (minima) for G_P (G_{AP}). Vice versa, negative local moments for odd Cr layers cause a local maxima (minima) for G_{AP} (G_P).

The periodic maxima and minima of G_P and G_{AP} cause a pronounced even-odd effect with periodic changes of $G_P > G_{AP}$ and $G_P < G_{AP}$. Consequently, this results in an oscillation of the TMR ratio shown in Fig 4a. This oscillation with a period of 2-ML is connected to a periodic sign reversal of the TMR ratio.

The TMR ratio with no Cr spacer is about 6800%. This order of magnitude shows up again only for a Cr thickness of 2 monolayers. In particular we would like to emphasize that the TMR value of approximately 8200% is larger than that one found for Fe/MgO/Fe MTJs without any Cr interlayers. The TMR ratio for 1 ML is strongly reduced about two orders of magnitude. Apart from the large amplitude for 2 ML, this reduced level is reached and maintained – alternating between about plus-or-minus 100% – for all thicker Cr films.

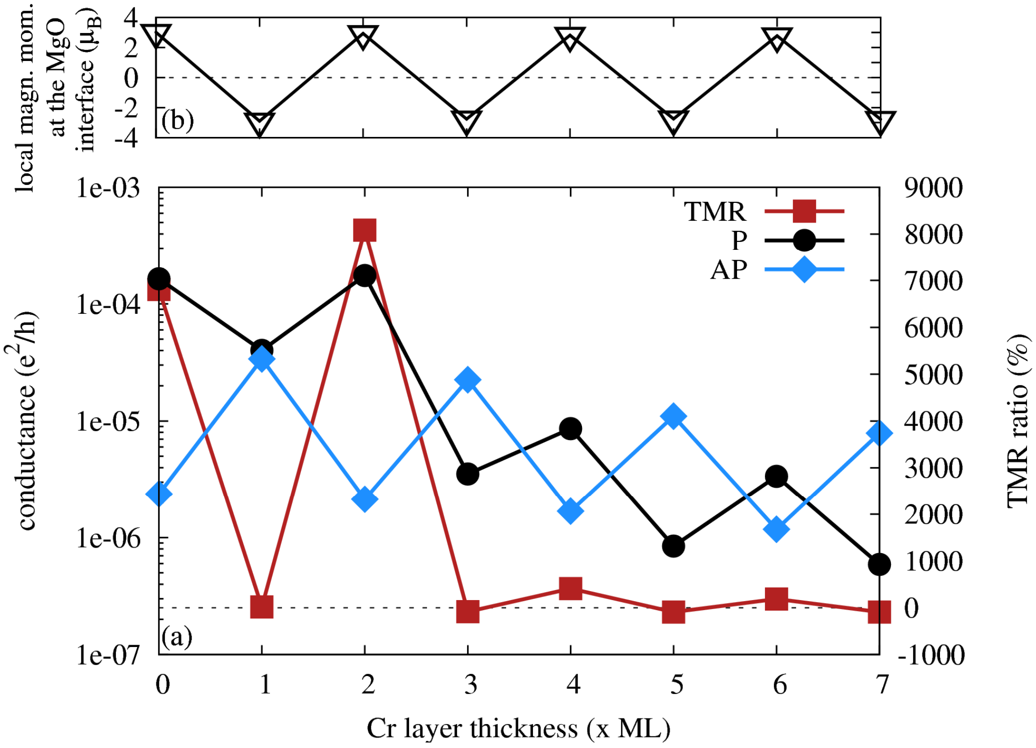

Figure 4. (a) As Figure 3, but for Fe/*x*(Cr)/MgO/Fe versus Cr thickness. (b) Local magnetic moment of the magnetic layer at the interface for each Cr thickness *x*.

CONCLUSIONS

Bcc Co interlayers at both interfaces of Fe/MgO/Fe junctions do not *per se* improve TMR ratios as compared to Fe/MgO/Fe junctions. Only specific Co thicknesses, namely 3 and 5 ML, result in larger TMR ratios.

The insertion of a single, layer-wise antiferromagnetic Cr buffer causes 2-ML oscillations of the conductances as a function of the Cr thickness, which show up as an even-odd change of the TMR's sign. The TMR ratio is generally, compared to the case with no Cr spacer, reduced about two orders of magnitude. Only a 2 ML thick Cr spacer is found to reproduce a sizeable larger TMR ratio.

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