Effect of submonolayer MgO coverages on the electron-spin motion in Fe(001): Experiment and theory

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It is shown that the spin polarization direction of the reflected electrons on Fe(001) strongly changes with minute amounts of MgO. Our *ab initio* electronic band structure and spin-dependent electron reflection calculations reveal that the MgO-induced out-of-plane relaxation of the Fe surface layer is responsible for this behavior. Our study points toward the subtle feature that the major change of the spin-dependent electron reflection properties of the Fe(001) surface is already caused by the very first MgO coverage.

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The interface system MgO/Fe(001) has attracted great interest in recent years for mainly two reasons: first, very large room temperature tunnel magneto-resistance (TMR) has been predicted and observed in epitaxial Fe/MgO/Fe(001) magnetic tunnel junctions (MTJs).[1,2] Second, magnetization switching by spin-transfer torque (STT),[3–5] being one of the most intriguing new concepts of contemporary magnetism,[6,7] and the behavior of spin-torque oscillators,[8,9] have been studied in Fe/MgO/Fe(001) and related MgO-based MTJs. In both cases, the MgO/Fe interface and in particular its spin-dependent electron reflection properties are of great importance, as they determine the electron transport polarization in TMR experiments as well as the transferred spin angular momentum in STT experiments. For instance, it is explored from various studies that interface oxidation influences the TMR values.[10] These findings spurred intense experimental and theoretical efforts to elucidate the influence of MgO on the Fe(001) surface and its spin-dependent electron reflection properties. Although many results have been published concerning the interfacial structure of MgO/Fe[11–19] and its influence on both TMR and STT experiments,[2,20] little effort was devoted to elucidate the spin-dependent electron reflection properties of MgO/Fe interfaces. It is thus an open question how the spin-dependent electron reflection properties of the Fe(001) surface are modified by MgO coverage.

In the attempt to unravel the interfacial spin-dependent reflection properties of MgO/Fe(001), the knowledge of the motion of the electron-spin polarization upon reflection from the interface is needed. This information can be drawn from spin-polarized electron reflection experiments, in which spin-polarized incoming electrons are spin-analyzed after reflection from the interface. Here, we report such experiments. For certain electron energy ranges the effect of minute amounts of MgO on the motion of the spin polarization is drastic. We assume that a strong out-of-plane relaxation of the Fe surface layer, which is induced by MgO, is at the origin of this behavior. This is corroborated by *ab initio* electronic structure calculations by means of the Korringa-Kohn-Rostoker (KKR) method.[21]

The origin of the motion of the electron-spin polarization in ferromagnetic films is the spin-dependent scattering of electrons within the film and at its surface.[22,23] Supposing a completely spin-polarized electron beam with its initial spin polarization $\mathbf{P_0}$ perpendicular to the magnetization $\mathbf{M}$ of the film, the spin part of the incident electron wave function is described by a coherent superposition of a majority-spin $\chi^{\uparrow}$ and a minority-spin $\chi^{\downarrow}$ wave function (with their moments parallel and antiparallel to $\mathbf{M}$, respectively) with equal amplitudes. Because of spin-dependent scattering, the spin wave function of the beam after reflection from the film reads: $\chi \propto |r^{\uparrow}|\exp(i\theta^{\uparrow})\chi^{\uparrow} + |r^{\downarrow}|\exp(i\theta^{\downarrow})\chi^{\downarrow}$ with $|r^{\uparrow,\downarrow}|$ and $\theta^{\uparrow,\downarrow}$ are, respectively, the moduli and the phases of the spin-dependent reflection amplitudes. This change of the spin wave function corresponds to a precession of $\mathbf{P}$ around $\mathbf{M}$ by an angle $\varepsilon = \theta^{\downarrow} - \theta^{\uparrow}$ and a rotation by an angle $\phi = \arctan[(|r^{\uparrow}|^2 - |r^{\downarrow}|^2)/2|r^{\uparrow}||r^{\downarrow}|]$ in the plane spanned by $\mathbf{P}$ and $\mathbf{M}$,[24] with $|r^{\uparrow,\downarrow}|^2 = R^{\uparrow,\downarrow}$ the spin-dependent reflectivity (see Fig. 1).

To understand the existence of the spin motion at a simple level we assume that the electrons within the ferromagnetic layer experience an exchange interaction which shifts the states in the spin-down band higher in energy than in the spin-up band. In this simple model, the electronic structure near the surface can be described by a simple scattering potential in which the electron scatters from a rectangular potential that has different heights for spin-up and spin-down electrons. This elementary problem can be solved exactly, and one can therefore determine the reflected and the transmitted part of the wave function.[25] This simple calculation shows in fact the existence of spin precession and rotation in reflection (as well as in transmission). However, this simple model is not realistic because it does not include the details

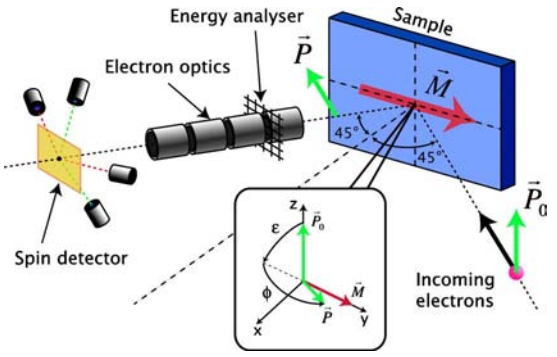

FIG. 1. (Color online) The experiment consists of a spin-polarized electron source, a sample that is magnetized remanently, a retardation grid for the energy analysis and a spin detection system. The two types of spin motion, i.e., a precession ε and a rotation ϕ, are shown.

of the electronic band structure. To be more realistic one has to calculate the spin-polarized low-energy electron diffraction from a Fe(001) surface as we did in our calculations.

The experiment consists of a spin-polarized electron source, an Fe(001) film which is magnetized remanently in-plane by a magnetic field pulse, and a spin detector (see Fig. 1). The 70% polarized electron beam is obtained from a GaAs-type source by optically pumping the crystal with circularly polarized light.[26] The beam is incident at 45° with respect to the sample surface with the in-plane projection of the wave vector along the [100]-direction of the film. To observe a maximum spin motion $\mathbf{P_0}$ has to be oriented perpendicularly with respect to $\mathbf{M}$ of the film.[22,23] It is only in a noncollinear geometry that $\mathbf{M}$ can exert a torque on the spin-polarization vector. Upon reflection from the sample, the specular beam passes through a retarding field energy analyzer. The spin polarization of the elastically scattered electrons, to which we restrict our discussion, is finally measured by a Mott detector, which exploits the left-right asymmetry of electron scattering due to spin-orbit interaction.[27] To get rid of any experimental asymmetry (for instance due to a misalignment of the beam of scattered electrons with respect to the Mott detector), the direction in space as well as the relative alignment of $\mathbf{P_0}$ and $\mathbf{M}$ must be interchanged. On reversing $\mathbf{P_0}$, only ϵ changes sign, while on reversing $\mathbf{M}$, the sense of both precession and rotation change sign. By combining thus properly the four different measurements the values of both spin motion angles are obtained. This technique also eliminates any effect resulting from spin-orbit interaction.

In a first step, Fe(001) films of 40 ML thickness are deposited at room temperature from an Fe rod heated by electron-beam bombardment on an Ag(001) single crystal. In a second step, MgO was deposited at room temperature at a rate of about 0.1 ML/min from pieces of stoichiometric MgO single crystals by electron-beam bombardment. All thicknesses are determined by a quartz microbalance. The oscillatory behavior of the reflection high-energy electron diffraction (RHEED) specular intensity as a function of MgO coverage [see inset in Fig. 2 (top)] shows that MgO grows on Fe(001) in a layer-by-layer fashion at least up to a thickness of 7 ML. Earlier STM work showed that MgO films of a few MLs are uniform in thickness with a quite small rms roughness.[28] We note that MgO films being thermally evaporated under ultrahigh vacuum conditions from stoichiometric MgO are known to be nearly stoichiometric or slightly oxygen deficient.[29] To exclude a possible change of the surface magnetism during MgO deposition, a thin Fe film of 6 ML thickness has been prepared and measured by the magneto-optical Kerr effect as a function of MgO coverage. In particular in the submonolayer coverage regime, we observe no significant changes of the saturation magnetization, the coercive field, and the remanence. In particular, no change of the easy direction of magnetization has been found. Moreover, both experiments[13] and calculations[30] have shown that the Fe magnetic moment at the MgO/Fe interface is rather enhanced (by about 35%) than reduced. Our own calculations, showing a magnetic moment of $3\mu_B$ at the interface, i.e., an enhancement of 36%, are in very good agreement with these findings.

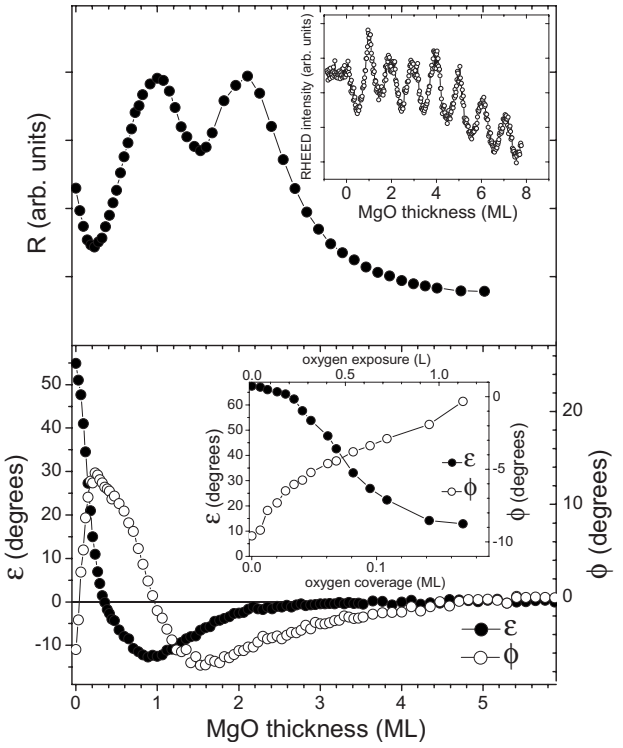

FIG. 2. (Top) Spin-averaged electron reflectivity R as a function of MgO coverage. The inset shows the RHEED intensity as a function of the MgO coverage. (Bottom) Precession angle ε and rotation angle ϕ as a function of MgO coverage. The inset shows ε and ϕ as a function of the oxygen coverage (bottom scale) and the exposure in Langmuir (L; $1\mathrm{L}=10^{-6}$ Torr·s) (top scale). The data of Sakisaka *et al.*,[32] which provide a relation between the exposure and the coverage, were used to translate exposures into coverages. The primary electron energy is $E-E_F=7$ eV. The Fe film thickness is 40 ML.

Figure 2 (top) shows the spin-averaged electron reflectivity (R) as a function of the MgO thickness at a primary electron energy ($E-E_F$) of 7 eV. Two intensity maxima with ML-periodicity (1 ML=0.22 nm) can be identified and attributed to periodic variations of the film morphology alternating between filled and incompletely filled atomic layers. Figure 2 (bottom) shows ε and ϕ as a function of MgO coverage for $E-E_F$ of 7 eV. Already 0.15 ML of MgO are sufficient to halve ε and even change sign of ϕ. Interestingly, ϕ approaches zero less rapid than ε. For coverages above 0.5 ML both quantities are 90° out-of-phase, i.e., ϕ exhibits its strongest change when ε is in its minimum and vice versa. We attribute the structures at 0.9 ML for ε and at 1.5 ML for ϕ to the appearance of a quantum interference in the MgO film.[31] As we find a drastic effect on ε and ϕ for very small coverages, we focus therefore on the range below 1 ML.

Figure 3 shows ε (top) and ϕ (bottom) as a function of $E-E_F$ for different MgO coverages. For small coverages (up to 0.18 ML) we find different behaviors depending on the energy range. While a strong reduction of ε with MgO coverage is found in the energy range from 7 to 9 eV and from 32 to 38 eV, a relatively strong increase is observed between 13 and 17 eV. For all other energies the changes are relatively small. In the case of ϕ the situation is quite similar

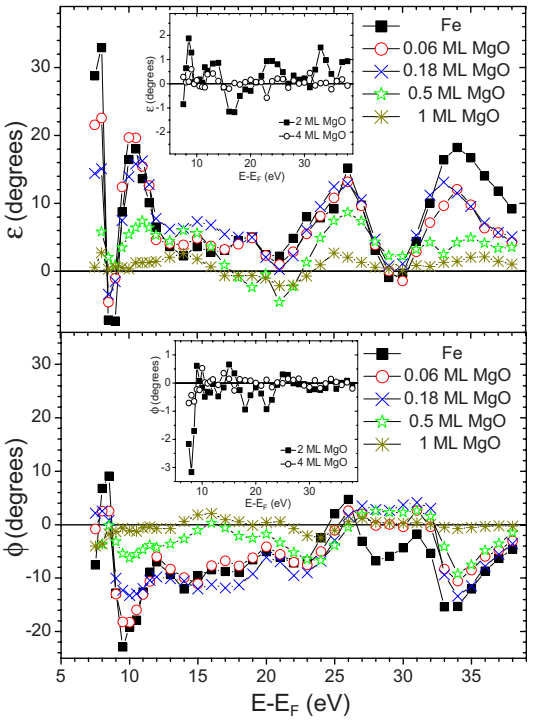

FIG. 3. (Color online) (Top) Precession angle ε and (bottom) rotation angle ϕ as a function of the primary electron energy for different MgO coverages. The Fe film thickness is 20 ML.

with the exception that between 27 and 31 eV ϕ is strongly reduced, while ε does not change significantly. Finally, for MgO coverages larger than 4 ML both spin-motion angles are practically zero for all energies (see insets in Fig. 3). This is due to a small electron inelastic mean free path of about 2–3 ML of MgO in this energy range.

Since a strong modification of the magnetization is excluded, we assume that the origin of this strong sensitivity of the spin-motion angles on the MgO coverage is an out-of-plane relaxation (expansion) of the Fe surface layer induced by MgO which is much stronger than the out-of-plane relaxation (compression) of about −1% which exists already for the uncovered Fe surface.[33] This in turn results in a change of the Fe electronic structure leading to a change of the spin-dependent reflection properties. In the simple potential-step model which we presented before this might correspond to a change of the spin-dependent potential step heights. In fact, surface x-ray diffraction experiments found that the first Fe interlayer distance is expanded up to 18% relative to the bulk value (0.143 nm) due to MgO coverage.[12] Most importantly, already a submonolayer coverage of 0.35 ML induces a significant relaxation (of 10%). Furthermore, the same experiments gave also clear evidence for the presence of a substoichiometric Fe-O layer between the Fe substrate and the MgO layers and it is this Fe-O layer which is believed to be responsible for the strong expansion of the first Fe interlayer distance. This leads us to question whether oxygen coverage of the Fe surface alone would lead to a similar behavior of the spin motion. Indeed, measurements at an electron energy of 7 eV, for which in the case of MgO strong changes are seen, show that both spin-motion angles exhibit also a strong decrease with oxygen coverage [see inset in Fig. 2 (bottom)].

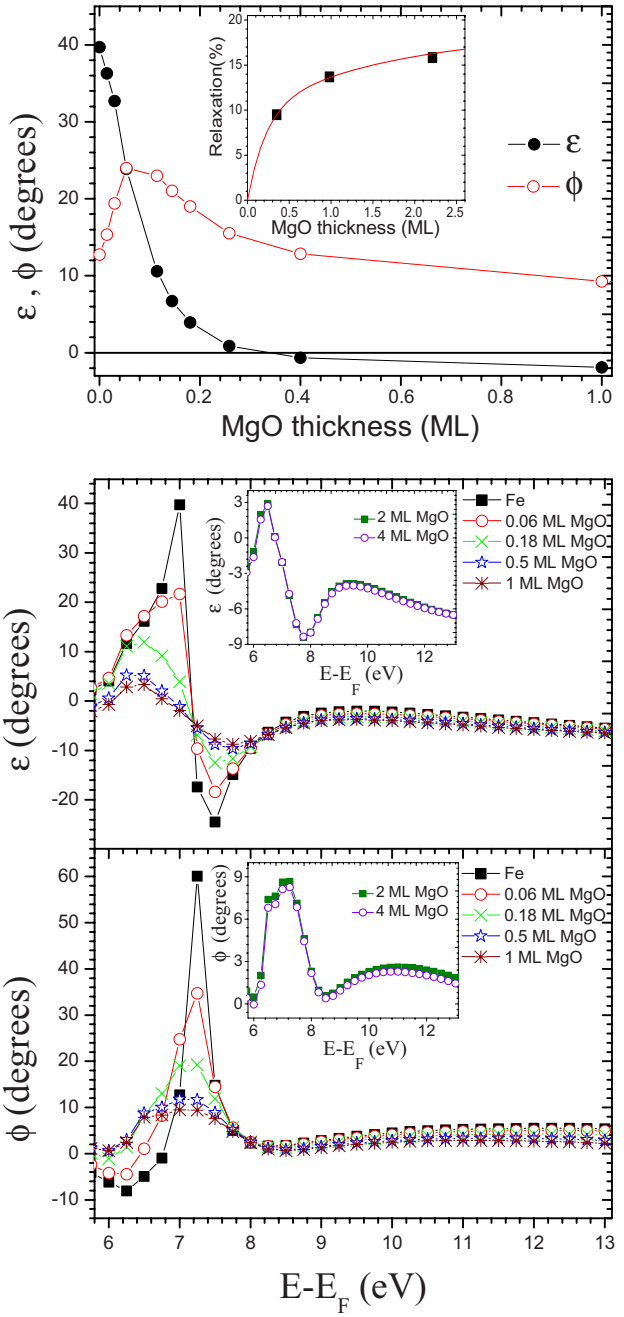

FIG. 4. (Color online) (Top) Calculated ε and ϕ as a function of MgO coverage for $E-E_F=7$ eV. The inset shows the relation (line) between the degree of relaxation and the MgO thickness, obtained by fitting an exponential to the experimental data (dots) of Meyerheim *et al.*[12] (bottom) Calculated ε and ϕ as a function of the primary electron energy for different MgO thicknesses.

Already ~0.1 ML of oxygen is sufficient to halve both ε and ϕ. This strongly suggests that the O-Fe bonds within the surface layer, which also lead to an out-of-plane relaxation of the Fe surface layer,[34] determine the spin-dependent reflection properties. We emphasize that for such small oxygen coverage the magnetization of Fe(001) films is not influenced.[35]

From the preceding we have learned that the electron spin-motion angles depend strongly on MgO coverage and might be explained by the out-of-plane relaxation of the Fe

surface layer. To prove this assumption we performed calculations for uncovered Fe in which the out-of-plane lattice constant of the surface layer has been varied.[36] We emphasize that the calculations were focused on the most prominent spectroscopic structure in the energy range from 7 to 9 eV. For higher energies no comparison is made with theory because due to the limited wave function basis-set of the linear muffin-tin orbital (LMTO) method[37] the determination of the electronic structure at higher energies is not very reliable. We emphasize, however, that the LMTO method was used only to provide the self-consistent potential of the relaxed Fe surface. - We have then taken the converged potentials and used them to compute the spin motion upon reflection using the Green's function formalism within the Korringa-Kohn-Rostoker (KKR) method.[21]

To compare the calculations with the experimental data as a function of MgO coverage, the x-ray data of Meyerheim *et al.*,[12] which provide a relation between the MgO thickness and the strength of the out-of-plane relaxation [see inset in Fig. 4 (top)], were used to translate the values of the out-of-plane relaxation in our calculations into MgO thickness values. Figure 4 (top) shows the calculated spin-motion angles as a function of MgO coverage, while the bottom figures show them as a function of primary electron energy for different MgO thicknesses. By comparing Fig. 4 with the experimental data in Figs. 2 and 3 we note a qualitative agreement with experiment. This leads us to the conclusion that the change of the Fe interlayer distance is indeed responsible for the behavior of the electron-spin motion upon reflection. We emphasize that better agreement cannot be expected, since there is no scattering at MgO in the model calculations. The effect of MgO is taken into account only via the relaxation of the surface Fe layer. Consequently, also the effect of quantum-well states in the MgO layer as well as that of the inelastic mean free path of the electrons in MgO are ignored in the calculations.

In conclusion, the interface system MgO/Fe(001) is studied by spin-polarized electron reflection experiments as a function of the MgO coverage as well as a function of the primary electron energy. A very strong sensitivity of the spin-motion angles ε and ϕ on the MgO coverage is observed for certain energy ranges. Magneto-optical Kerr effect experiments did not show any strong change of the Fe surface magnetism during MgO deposition, and therefore no such effect is at the origin of these strong variations of the spin-motion angles. Indeed, the qualitative agreement of our *ab initio* calculations with the experimental data suggests strongly that the out-of-plane relaxation of the Fe surface layer, induced by MgO, is responsible for this behavior. The present findings underline the importance of details of the interfacial structure for the spin-dependent reflection properties in the system MgO/Fe.

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