

Surface x-ray diffraction analysis of the MgO/Fe(001) interface: Evidence for an FeO layer

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Using surface x-ray diffraction we have investigated the geometric structure of the interface between thermally grown MgO layers and Fe(001). The MgO/Fe(001) interface is part of the Fe/MgO/Fe junction, which has become a prototype system in the study of the tunneling-magnetoresistance (TMR) effect. For all samples studied in the MgO coverage range between about 0.35 and 4.6 ML we find clear evidence for the presence of a substoichiometric FeO layer between the bulk Fe crystal and the MgO adlayers. The partial oxidation of the Fe(001) surface takes place during deposition of the first MgO monolayer and approaches a concentration limit, where about 60% of the Fe(001) hollow sites are occupied by O ions. The formation of a bulklike sixfold-coordinated Mg coordination at the MgO/O/Fe(001) interface might be accounted for stabilizing the interface structure, in which several Fe-O distances are strained by up to (10% with respect to their bulk analog. The presence of the strained interfacial FeO layer is likely to have considerable consequences on the magnitude of the TMR effect.

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I. INTRODUCTION

Since the first tunnel-magnetoresistance (TMR) measurement on a ferromagnetic-insulator-ferromagnetic (FM-I-FM) trilayer junction in 1975,¹ the preparation and characterization of tunneling devices has become a major research field.^{2-4,6-8} This is because of their importance for possible technological applications such as the development of magnetic random access memory (MRAM) devices. So far most attention has been focused on the investigation of TMR devices composed of amorphous oxide barriers like corundum (Al₂O₃). From the experimental point of view, the growth of an amorphous Al₂O₃ layer is comparatively easy as compared to the epitaxial growth of a single-crystalline barrier. In contrast, their electronic and structural properties are difficult to characterize. In devices containing an amorphous barrier, electron tunneling is dominated by random hopping between oxide resonance states induced by defect sites, leading to the randomization of the parallel component of the electron momentum (k_{\parallel}). Consequently, all properties related to the oxide band structure are averaged out and the magnitude ($\Delta R/R$) of the TMR effect can simply be described by the effective spin polarization of the ferromagnetic films. As a result of the structurally poorly characterized barriers and interfaces there is also a considerable scatter in the published values for $\Delta R/R$. Nevertheless, high values of up to 27% (Ref. 6) were reported for Co/Al₂O₃/Ni₈₀Fe₂₀ junctions by Moodera *et al.* at 77 K in agreement with Julliere's model.

As a consequence of the experimental and theoretical problems encountered with amorphous oxide barriers, single-crystalline oxide barriers such as MgO appear to be primary candidates for the preparation of TMR junctions, since they offer the opportunity to control their structural and physical

properties in a more reproducible way. Furthermore, they are directly accessible by theory because electron tunneling is ballistic in this case; i.e., k_{\parallel} is conserved. Huge values for $\Delta R/R$ of the order of several thousand percent were predicted in a recent theoretical study of Butler *et al.*⁹ for the Fe/MgO/Fe TMR junction using the Landauer formalism. The authors have given evidence for the importance of the symmetry relation between the propagating states in the Fe electrodes and the evanescent states in the MgO barrier. Their conclusion is that the tunneling magnetoresistance is due to the strongly peaked conductance in the majority channel at $k_{\parallel}=0$ for parallel alignment, whereas minority channel conductance is dominated by states associated with k_{\parallel} values close to interface states. From their results it can be concluded that a reliable comparison between experiment and theory relies on the detailed knowledge of the interface structure. Structural defects strongly influence the electron tunneling in both the majority and minority channels. For example, the modification of interface states can result in a less peaked conductance around $k_{\parallel}=0$ and reduce considerably the magnetoresistance amplitude $\Delta R/R$. Consequently, a detailed understanding of the metal/oxide interface structure is a prerequisite to determine the role of interface states on the TMR effect.

In contrast to these promising properties of monocrystalline barriers there are only a few attempts to grow and to characterize the corresponding TMR junctions. The prototype character of the Fe/MgO/Fe junction is based on the fact that MgO can be grown lattice matched on Fe(001) (Refs. 10 and 11) either by electron beam deposition or by pulsed laser deposition (PLD). The low lattice mismatch (3.8%) between Fe and MgO and the large difference of the surface free energies of Fe (2.9 J/m²) and MgO (1.1 J/m²) (Refs. 14 and

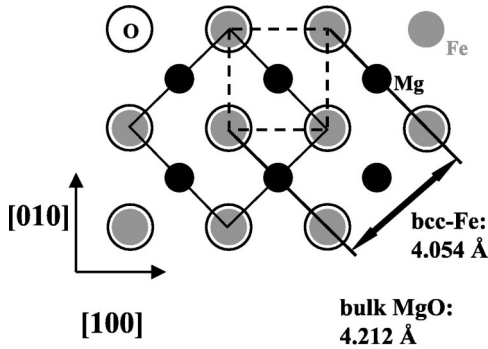


FIG. 1. Traditional structure model (without FeO-like interface layer) of the MgO/Fe(001) interface in top view. Only the first MgO layer is shown. The surface Fe atoms are represented as solid gray circles. Mg ions of the first MgO layer are shown as black circles. The O ions (large open circles) are located on top of the Fe atoms. The dashed and solid squares correspond to the surface unit cells of the bcc Fe substrate and the face-centered lattice of the MgO adlayer, emphasizing the mutual 45° orientation between the lattices.

15) allow the growth of single-crystalline layers. Although first attempts to prepare a crystalline Fe/MgO/Fe(001) junction using a MgO(001) crystal were not successful because of pinholes leading to an Ohmic contact,¹⁶ recent experiments depositing MgO by electron beam deposition on a Fe(001) whisker were able to demonstrate ballistic tunneling at room temperature,^{17–19} although due to equipment limitations the TMR effect could not be measured. In summary, one can state that a detailed investigation of the interface structure is needed to understand the TMR properties.

Previous studies^{10,11,20} only investigated the epitaxial relationship between MgO and Fe (MgO[100] parallel to Fe[110]), the layer-by-layer growth mode of MgO on Fe(001) up to about five monolayers (ML) of thickness, and the monocrystallinity of MgO(001) by monitoring low-energy electron diffraction (LEED) spots. Some information on the geometric structure was provided by a quantitative LEED study only for the inverse interface [Fe/MgO(001)] proposing that the Fe atoms adsorb on top of the O atoms at a distance of about 2.0 Å.²⁰ Figure 1 shows the so far generally assumed MgO/Fe(001) interface structure, which was also considered by Butler *et al.* in their TMR calculations.⁹ The gray circles represent the surface Fe atoms, the Fe-surface unit cell is indicated by the dashed square. The small solid and large open circles correspond to the Mg and O ions, respectively. Only the first MgO monolayer is shown; subsequent layers are stacked over the preceding ones by placing the O (Mg) ions on top of the Mg (O) ions. The face-centered unit cell of the first MgO adlayer is indicated by the solid square emphasizing the mutual 45° orientation between the Fe and MgO surface unit cells. The MgO lattice is laterally contracted by 3.8% relative to the bulk to accommodate to the Fe lattice (4.054 Å vs 4.212 Å).

At some variance to this model our recent surface x-ray diffraction (SXRD) study has proved that this model is not correct in detail.²¹ It could be shown that the first MgO layer is separated from the bulk Fe substrate by a substoichiometric FeO layer, in which the O occupation is about 0.6 ML; i.e., 60% of the Fe(001) hollow sites are occupied by O ions.

This interfacial layer is expected to considerably influence the tunneling properties of the junction. Preliminary calculations by Zhang and Butler²² taking into account a complete FeO layer indicated a TMR magnitude of 76% at 0 K. This appears to be a more realistic value when compared with experimental values reported so far. In the present paper we focus on the MgO-coverage-dependent x-ray structure analysis of the MgO/Fe(001) interface, which indicates strained interatomic distances across the interface and show that the oxidation of the Fe(001) surface takes place during deposition of the first MgO monolayer.

II. EXPERIMENT

The experiments were carried out at the wiggler beamline DW12 of the storage ring LURE in Orsay (France) using a six-circle ultrahigh-vacuum (UHV) diffractometer. A single-crystalline Fe(001) substrate crystal ($\varnothing=9$ mm) was prepared in UHV by standard procedures until only traces of nitrogen were detectable by Auger electron spectroscopy (AES). LEED showed a bright well-contrasted (1×1) pattern, also indicating the cleanliness of the surface, which is known to exhibit a $c(2\times 2)$ superstructure even in the case of minor C or S contamination. MgO was deposited *in situ* by electron beam evaporation from a polycrystalline rod. During deposition the pressure rose from 2×10^{-10} mbar to 1×10^{-9} mbar. Vassent *et al.*^{12,13} relate this to molecular oxygen generated by recombination of atomic oxygen issued from the MgO source at the walls of the UHV chamber. In their studies they also showed by using mass spectrometry and thermodynamic considerations that it is atomic O and Mg which is released from the MgO source and that an excess of O relative to Mg arrives at the sample. From their results the oxidation of the Fe(001) surface can be anticipated. In this context it should be noted that the structure of O/Fe(001) was studied previously,^{5,23,24} however, in these experiments the deposition was carried out by dosing with molecular oxygen.

In total, seven data sets at six different MgO coverages between 0.35 and about 4.6 ML were collected. After each deposition, the coverage calibration was carried out using AES and cross-checked afterwards in comparison with the results of the SXRD analysis. In general, there is agreement between the AES- and SXRD-derived coverages within 5–15%. Integrated x-ray intensities were collected using a wavelength of $\lambda=0.887$ Å by transverse scans under total reflection conditions of the incoming beam. The high sample quality is confirmed by the transverse full width at half maximum of the antiphase (0 1 0.1) crystal truncation rod (CTR) reflection of 0.1° , which corresponds to the mosaic spread of the sample. After correcting the measured intensities for active sample area and Lorentz and polarization factors,^{25,26} for each data set up to 125 symmetry-independent structure factor amplitudes $|F|$ along the (10*l*), (11*l*), (20*l*), and (21*l*) CTR's were derived. The standard deviations (σ) of the $|F|$ values were estimated from the reproducibility of symmetry-equivalent reflections (two for each independent reflection) and the counting statistics as outlined in Ref. 27. In general,

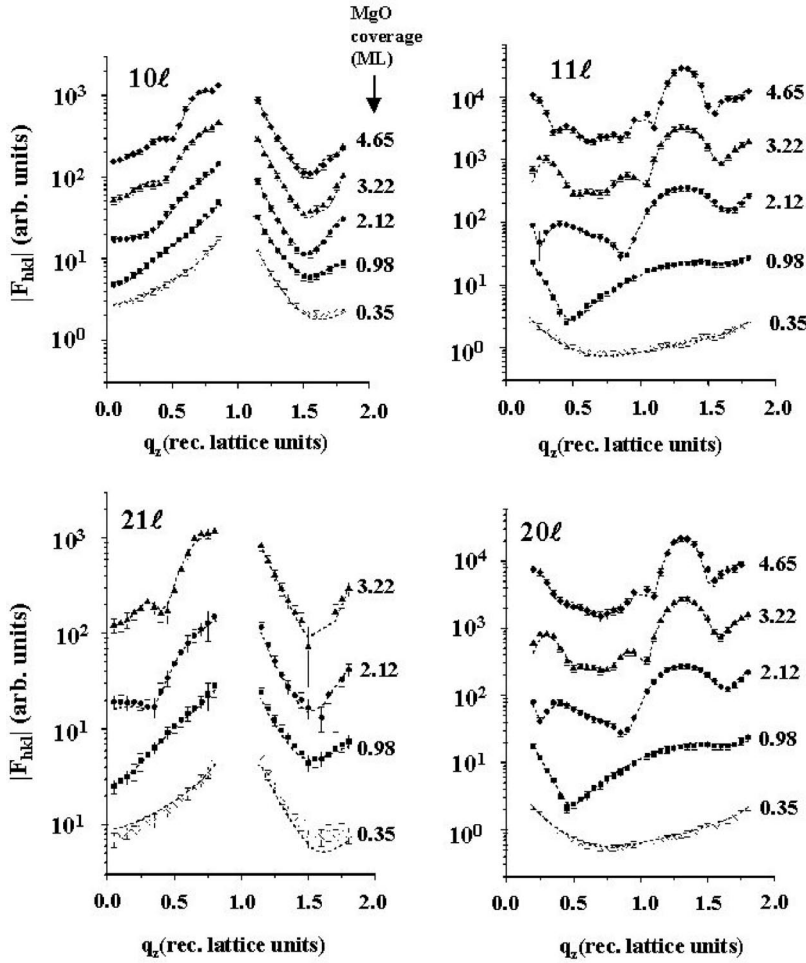


FIG. 2. Measured (symbols) and fitted (lines) structure factor amplitudes along the $(10l)$, $(11l)$, $(20l)$, and $(21l)$ crystal truncation rods for MgO/Fe(001). The individual rods are shifted vertically for clarity. The numbers at the curves indicate the MgO coverage in monolayers.

σ is in the 3–5 % range, a value which can be considered as quite good for SXRD data.

The CTR's arise due to the truncation of the crystal and can be treated theoretically by calculating the semi-infinite sum over the crystal lattice planes along $[001]$. Neglecting absorption, the CTR structure factor amplitude F_{hkl} of the bcc Fe crystal covered with adsorbate atoms (i) at positions (x_i, y_i, z_i) with respect to the Fe-surface unit cell and a fractional occupancy Θ_i is given by

$$|F_{hkl}| = \left| \frac{f_{Fe}}{1 - e^{-i\pi(h+k+l)}} + \sum_i \Theta_i f_i e^{i2\pi(hx_i + ky_i + lz_i)} \right|. \quad (1)$$

The first and second terms correspond to the bulk truncated substrate and adsorbate scattering contribution, respectively. The f_i 's are the atomic scattering factors of the atoms. A more rigorous analysis²⁸ shows that l is a continuous parameter, whereas h and k are integers. The CTR's are peaked at the bulk Bragg conditions $h+k+l=2n$ (n integer), but are weak in between. At the antiphase conditions $(h+k+l)=2n+1$, the structure factor of the uncovered bulk truncated Fe(001) surface equals $f_{Fe}/2$; i.e., the scattered intensity is equivalent to 1/4 of that corresponding to 1 Fe ML. Thus,

ordered surface adsorption in general strongly modifies the CTR intensity, which allows one to carry out a surface-sensitive structure analysis.

III. RESULTS

The solid symbols in Fig. 2 represent the measured structure factor amplitudes $|F_{hk}^{obs}(q_z)|$ along the $(10l)$, $(11l)$, $(20l)$, and $(21l)$ rods after depositing 0.35, 0.98, 2.12, 3.22, and 4.65 ML MgO. The different curves are shifted vertically for clarity. Direct inspection of the intensity distribution along the rods allows some general conclusions. First, the CTR's show a rapid modulation along q_z , indicating the presence of several adlayers. The increasing number of maxima and minima between the bulk Bragg reflections [at $l=0,2$ for the $(11l)$ and $(20l)$ rods and at $l=1$ for the $(10l)$ and $(21l)$ rods] is due to the increasing number of MgO adlayers. Further, from the identical profiles of the $(10l)$ and $(21l)$ rods as well as of the $(11l)$ and $(20l)$ rods only high-symmetry adsorption sites such as $(x, y) = (0, 0)$ and $(1/2, 1/2)$ within the Fe(001) surface unit cell can be inferred, since for these rods the lateral component of the scattering phase, $\exp[i2\pi(hx + ky)]$, is identical. So far, this is compatible with the “traditional” model of the MgO/Fe(001) interface in which MgO directly grows on Fe(001) as outlined in Fig. 1.

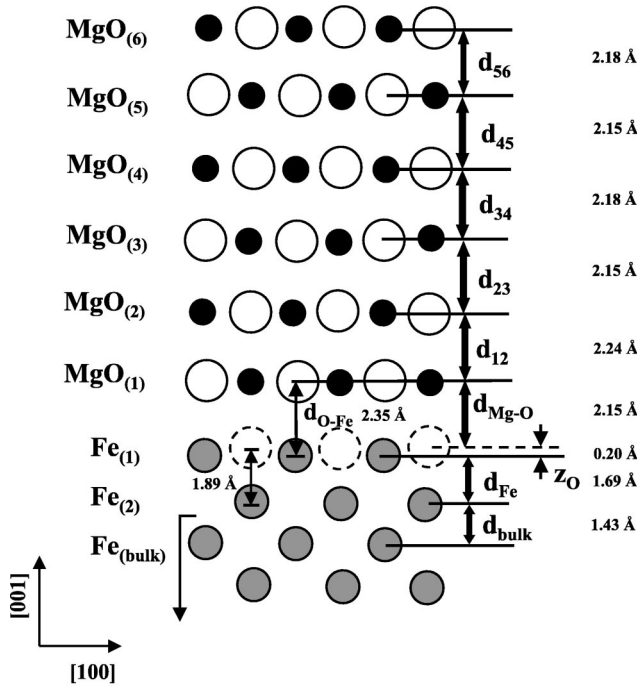


FIG. 3. Side view of the MgO/Fe(001) structure model as derived from the x-ray analysis. In addition to the Fe atoms and the MgO layers ($i = 1, \dots, 6$) there is a fraction of O ions (large dashed circles) in the hollow sites of the Fe(001) surface. The labels refer to Table I. The indicated distances refer to the 4.65 ML sample.

For refining this structure model, several fit parameters were used. Apart from allowing for variations of the vertical distances between the MgO layers, between the first MgO layer and the Fe substrate and between the first and second Fe layers, the occupancy factors for the MgO layers were varied. In addition, one overall scale factor was used for all rods. The thermal Debye parameter ($B = 8\pi^2\langle u^2 \rangle$, where $\langle u^2 \rangle$ is the mean-squared displacement amplitude) was kept constant at $B = 0.6 \text{ \AA}^2$ for all atoms of the structure. Only for the low-coverage sample (0.35 ML) was the refinement of the B factors tried as will be discussed below.

On the basis of this model the unweighted residuum R_u (Ref. 29), is in the range of 10%, which—given the average standard deviation σ_{hkl} of the $|F_{hk}^{obs}(q_z)|$ values in the order of 3–5%—means that the deviation between $|F_{hk}^{obs}(q_z)|$ and $|F_{hk}^{calc}(q_z)|$ is about two standard deviations on average. This strongly indicates that the model is not correct in detail. By contrast, a satisfying fit (R_u in the range between 4.2% and 6.5%) is obtained by additionally placing O ions into the hollow sites of the Fe(001) surface. In this model only one extra position parameter—namely, the z position of the O ions—has to be varied. The high fit quality, which is achieved by introducing O ions, is evident by inspecting Fig. 2, where the dashed lines represent the calculated $|F|$ values on the basis of this model. The fitted $|F|$'s follow even the fine details of the measured CTR's.

In order to summarize the results of the structure analysis, Fig. 3 schematically shows the stack of layers at the interface between bulk Fe and the MgO layers. The interfacial O ions are sketched as large dashed circles; all other atoms are rep-

TABLE I. Structure parameters derived from the x-ray analysis for MgO/Fe(001) in the coverage range between 0.35 and 4.65 ML. The distances are given in \AA ; the error bars of the distance determination are in the 0.1–0.2 \AA range. The parameters refer to Fig. 3. Interlayer distances in bulk compounds: Fe: 1.433 \AA ; MgO: 2.106 \AA ; FeO: 2.145 \AA .

	d_{Fe}	d_{O-Fe}	d_{Mg-O}	z_O	d_{12}	d_{23}	d_{34}	d_{45}	d_{56}
Coverage									
0.35 ML	1.57	2.21	1.95	0.25					
0.98 ML	1.63	2.43	2.09	0.23	1.90				
2.12 ML	1.66	2.35	2.14	0.17	2.26	2.09			
3.22 ML	1.69	2.35	2.15	0.20	2.18	2.15	2.18		
4.65 ML	1.69	2.35	2.15	0.20	2.24	2.15	2.18	2.15	2.18

resented as in Fig. 1. The labels refer to Table I listing some interatomic and interlayer distances. Error bars of the distance determination are in the 0.10–0.15 \AA regime in general; however, for the 0.35 ML, sample they are in the 0.2–0.3 \AA range. This is due to the low contribution of the adatoms to the total scattering amplitude as a result of their low fractional occupancy.

The most remarkable result of this study is the presence of the (incomplete) FeO-interface layer, which neither in experimental nor in theoretical studies has been considered so far. In this context it must be emphasized that it appears difficult to detect the interfacial O layer with other surface analysis methods such as, e.g., x-ray photoelectron spectroscopy (XPS). XPS should in principle be able to identify a chemically shifted (partially oxidized) component related to the oxidized surface Fe atoms. However, the small shifted component must be resolved with respect to the large bulk metal contribution. Moreover, once measured another problem lies in the unambiguous assignment of the different components, because one could also infer some shift due to the interaction with the O ions of the first MgO layer.

In the following we discuss the geometric structure of the interface in some detail. As a consequence of the O adsorption on the Fe(001) surface, the first substrate interlayer distance (d_{Fe}) is expanded by up to 18% (1.69 \AA) over the bulk Fe interlayer distance (1.43 \AA). This can hardly be induced by MgO growth, since theoretical analyses^{9,30} have indicated a weak chemical interaction between Fe and MgO. Instead, the top Fe-interlayer expansion is related to the FeO formation at the Fe(001)-(1 \times 1) surface. Previous LEED and theoretical analyses on the oxidation of the Fe(001) surface have reported an expansion by 8% (Refs. 5 and 23) and 23% (Ref. 24), respectively. However, a direct comparison with our data needs a caveat, since Refs. 5 and 23 are concerned with a pure adsorbate structure, whereas in the present study the oxidized Fe surface is covered with MgO. There is evidence that this influences the interface structure as compared to the purely oxidized Fe(001) surface. For example, the adsorption height ($z_O \approx 0.20$) of the O atoms above the Fe surface (see Fig. 3) is lower than the corresponding value in the LEED analysis of the O/Fe(001)-(1 \times 1) structure by Jona and Marcus²³ (0.43 \AA). Thus, the interatomic O-Fe distances are comparatively short. For the 4.65 ML sample, which can

be regarded as quite representative, we determine nearest O-Fe distances of 2.04(15) Å (lateral) and 1.89(15) Å (vertical). These distances are somewhat low as compared to the corresponding FeO-bulk value (2.154 Å) but they are still in the range normally seen in other Fe-O structures (1.85–2.31 Å).³¹

The compression of the O-Fe distances might be explained by the presence of the MgO layer [labeled by MgO(1)] located directly above the interface O ions. First, in all samples the normal Mg-O distance (d_{Mg-O}) equals 2.15–2.20 Å (the 0.35 ML sample is a bit outstanding in this context, which we mostly attribute to the larger uncertainty of the distance determinations). This means that the vertical Mg-O distance closely corresponds to the bulk MgO value (2.106 Å). In contrast, the vertical O-Fe distance between the O ions of the first MgO layer and the Fe-surface atoms is expanded by about 10% over the corresponding bulk value. In Fig. 3 and Table I this distance is labeled by d_{O-Fe} . For d_{O-Fe} we determine values in the range between 2.35 and 2.43 Å (again with the exception of the 0.35 ML sample). Therefore, in order to keep the metal-oxygen distances (Mg-O and O-Fe) across the interface within reasonable limits, z_O must be in the range of about 0.2 Å. A larger z_O would imply an outward shift of the MgO layer to keep the Mg-O distance, which—given the already expanded Fe-O distance—appears hardly favorable. Finally, one could infer a rumpling of the first MgO layer of about 0.2 Å in order to adopt more closely the individual metal-oxygen distances at the FeO/MgO interface to the bulk values. To first order, a moderate rumpling should show up in an increased Debye parameter B of the corresponding atoms. However, refining this parameter as well as refining the z parameters of the individual atoms of the first MgO layer did not lead to improved fits in general. Some exception to this rule is the 0.35 ML sample, where we find enhanced Debye parameters for the interface O ions and the MgO layer [$B=2-5 \text{ \AA}^2$, corresponding to root-mean-square (rms) displacements $\sqrt{\langle u^2 \rangle}$ of the order of 0.15–25 Å]. This could be interpreted as an indication of the presence of some disorder; however, given the quite large error bars of the structural parameters characteristic for this particular sample, we do not think that these are reliable enough to allow definite conclusions.

We can summarize that the interatomic distances at the Fe/MgO interface are considerably strained. The complicated interface structure with its compressed and expanded distances might be accounted for the limited incorporation of O ions into the Fe(001) surface. The refined O-ion surface occupancy (Θ_O) is shown by the diamonds in Fig. 4 as a function of the MgO coverage. For the 0.35 ML sample we find $\Theta_O=0.45(10)$, which increases up to 0.75(10) for the 1.2 ML sample and is nearly constant at 0.60(10) at higher coverage. From these data it can be concluded that the oxidation of the Fe(001) surface takes place at the beginning of the MgO deposition ($\ll 1$ ML) and is completed after deposition of about 1 ML MgO. The 0.6 ML occupancy seems to be the limiting value for the given experimental conditions. Since theoretical analyses are not available so far, we may speculate that the formation and stability of the Fe(001)/FeO/

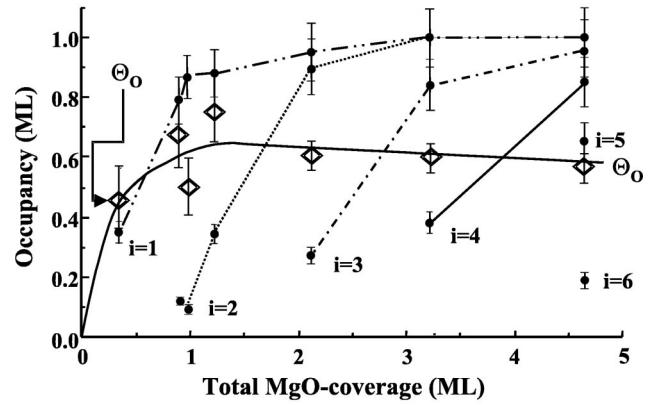


FIG. 4. Fractional occupancy of the individual MgO layers $i = 1, 2, \dots, 6$ plotted vs the total MgO coverage (circles). Successive filling of the layers with increasing MgO coverage indicates layer-by-layer growth. The diamonds represent the fractional occupancy of the interfacial O ions. The solid line is a guide to the eye emphasizing the saturation of the O occupancy at about 1 ML.

MgO interface is governed by a delicate balance of several competing contributions. Since the strain in the interface represents an “energy penalty” to the total free energy of the structure, which even might inhibit the energetically favorable (exothermic) full oxidation of the Fe(001) surface, one might question what could energetically stabilize the interface structure. The most important structural modification induced by the interface O ions is that the corresponding fraction of first layer of Mg ions experiences a bulklike sixfold O coordination instead of an incomplete fivefold coordination otherwise. A fivefold coordination of the Mg ions next to the Fe surface appears energetically unfavorable, possibly due to ionic repulsion. Stated differently, the Fe-O interface leads to a “smoother” transition from the bulk metal to the MgO layer. One parameter, which also could influence the O-surface concentration, is the O/Mg excess in the molecular beam. It was determined to be in the range between 1.3 and 2.3 for experimental conditions, which are quite close to ours in the present study.^{12,13} It might appear possible to avoid the FeO formation by decreasing the O/Mg excess, e.g., by simultaneously dosing with Mg.

Apart from the results directly related to the interface, the x-ray data also provide some details on the structure of the MgO layers. The MgO lattice is laterally contracted by 3.8% in order to accommodate to the Fe lattice, leading to an increase of the normal lattice constant c_0 . Taking from the refined interlayer distances d_{ij} (Table I) an average value of about 2.17 Å, the normal lattice constant equals about 4.34 Å (3% expanded over the bulk). This value is in good agreement with the expansion calculated by using continuum elasticity theory. The bulk-MgO Poisson coefficient (ν) equals 0.24.³² Inserting the lateral compression of $\epsilon_1 = \epsilon_2 = -0.038$ into the equation valid for cubic crystals, $\epsilon_3 / (\epsilon_1 + \epsilon_2) = -\nu / (1 - \nu)$, one obtains a normal expansion of $\epsilon_3 = +0.024$. The calculated 2.4% expansion corresponds to a lattice constant of $c_0 = 4.31 \text{ \AA}$, which is quite close to the value derived from our SXRD data.

Our x-ray analysis quantitatively confirms the layer-by-

layer mode which has been observed qualitatively on the basis of reflection high-energy electron diffraction (RHEED) oscillations and scanning tunneling microscopy (STM).^{17,18} The solid circles in Fig. 4 show the occupancy of the individual layers ($i = 1 - 6$). As observed in the STM images, we find that the layer-by-layer growth is not perfect and layer ($n + 1$) already grows when layer (n) is not fully completed. The error bars of the occupancy factors are in the range of about 15%; the two data sets taken at a MgO coverage close to 1 ML prove the reproducibility of the results. Similarly, the MgO layers exhibit a 1:1 stoichiometry within an error bar of about 10%.

Finally, our structure analysis points at a new direction for improving the Fe/MgO/Fe junction. So far it has been difficult to prepare smooth Fe layers on the MgO barrier. Topographic STM images of Fe on MgO show a considerably rough surface morphology corresponding to three-dimensional island growth (island diameter in the order of 5–10 nm) after thermal deposition of Fe on MgO.³³ This is due to the different surface free energies of Fe and MgO,^{14,15} which allows the layer-by-layer growth of MgO on Fe(001) but not vice versa. Based on the results of this work it appears tempting to introduce an FeO layer at the top electrode interface, Fe/MgO, creating a “mirror image” of the bottom electrode interface. First experiments depositing the first ML Fe on MgO under O(2) partial pressures in the 10^{-7} mbar range followed by 4 ML of Fe deposited under normal UHV conditions and annealed to 250 °C for 5 min resulted in a considerably smoother Fe-surface morphology. It is characterized by flat terraces separated by monoatomic steps.³³ One could speculate that this could be interpreted as being due to the formation of a FeO-like interface layer; however, a structure analysis has not been carried out so far.

IV. DISCUSSION

Apart from its implications on the stability of the interface structure, the FeO-interface layer is also expected to have significant influence on the electronic properties of the MgO/Fe junction and its related tunneling properties. So far it has been a very common approach in all calculations on TMR junctions to assume a direct transition of the metal to the insulating barrier. Our analysis shows that this is clearly not the case. Although our study directly only deals with the

MgO/Fe(001) interface, it seems plausible that different metal/barrier interfaces are not as “ideal” as generally assumed. Our results are important for first-principles calculations of Fe/MgO/Fe TMR junctions. These were carried out assuming a simple abrupt MgO/Fe(001) interface.⁹ They showed that the symmetry of the majority and minority states and the interface states of the minority electrons dominates the TMR. The minority interface states are expected to be strongly altered by any modification of the interface; consequently, the spin-dependent tunneling might be significantly changed as compared to that in an “ideal” interface. Preliminary calculations of Zhang and Butler²² using our structure model (but with a complete O-interface layer) are in favor of this assumption. A more realistic TMR of only 76% is found for $T=0$ K instead of several 1000% without the FeO layer.

In summary, we have analyzed the geometric structure of the interface between thermally grown MgO and single-crystalline Fe(001). The analysis of the x-ray intensities indicates the presence of a substoichiometric FeO-like interface layer between the Fe(001) substrate and the MgO layers. The oxidation of the Fe(001) surface takes place at the very beginning of the MgO deposition ($\ll 1$ ML) and reaches a limiting value, where about 60% of the Fe(001) hollow sites are occupied by O ions. Several metal-oxygen distances at the Fe(001)/FeO/MgO interface are strained [up to (10%)], which might limit the O concentration in the interface layer to 0.6 ML. The MgO layers grow in layer-by-layer mode; we find the MgO-interlayer distances strained by about +3% as a result of the in-plane contraction of 3.8% to accommodate the Fe(001) lattice parameter. This is in reasonable agreement with predictions from continuum elasticity theory. The presence of the FeO-like interface layer is likely to strongly affect the tunneling properties and the magnitude of the TMR effect. This appears not only important for the system investigated, but for metal/oxide interfaces in general.

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