

On the quality of molecular-beam epitaxy grown Fe/MgO and Co/MgO(001) interfaces

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We investigated in detail the structural, chemical, and magnetic properties of Fe at the interface with MgO. The samples were grown by molecular-beam epitaxy and analyzed by using reflection high-energy electron diffraction, scanning tunneling microscopy, Auger electron spectroscopy, x-ray photoemission spectroscopy (XPS), x-ray-absorption spectroscopy (XAS), and x-ray magnetic circular dichroism (XMCD) measurements. We discuss the presence or absence of C and O contaminants at the surface of the first electrode. We actually show that when growing the first Fe layer on the initial MgO substrate, a carbon contamination at the surface of the annealed Fe layer is observed leading to a C(2×2) surface structure. We propose a method to eliminate this carbon contamination and to get interfaces free of carbon. On the other hand, we observed that the reactivity of the Fe surface to oxygen contamination is greatly influenced by the presence of carbon or absence on the surface. The influence of oxygen adsorption at the Fe surface on the MgO growth will also be discussed. Concerning the hybridization between Fe or Co with MgO, XPS, and XAS-XMCD analyses clearly demonstrate that the charge transfer is weak, and that the magnetization at the interface is larger than in bulk ($3\mu_B/\text{at}$ for Fe). With such clean interfaces, reproducible magnetoresistance equal to 160% are obtained on Fe/MgO(3 nm)/Fe(001) $100 \times 100 \mu\text{m}^2$ tunnel junctions at room temperature in our laboratory. Finally, we show that pollution at the bottom Fe/MgO interface modify drastically spin-dependent tunneling properties. © 2006 American Institute of Physics. [DOI: [10.1063/1.2165914](https://doi.org/10.1063/1.2165914)]

I. INTRODUCTION

Magnetic tunnel junctions (MTJs) are of great interest due to their potential applications as read heads or magnetic sensors. These devices are composed of two magnetic electrodes separated by an insulator. They operate on the basis of the tunnel magnetoresistance (TMR) that is a spin-dependent tunneling effect. Numerous experimental and theoretical works show the key role of the nature of the bonding at the metal/insulator interface in the tunneling process.¹ Indeed, interfacial contaminants or impurities can strongly modify tunneling properties particularly for high symmetric systems such as epitaxial layers. Consequently, it becomes crucial to control the chemical quality of the interfacial area. The Fe/MgO/Fe(001) system is currently under scope since theoretical calculations predicted a very high TMR.² These calculations were performed for *perfect* interfaces. Nevertheless, the relevant point is to characterize *real* interfaces. For example, some calculations taking into account a possible FeO interfacial layer predict a dramatic decrease of the TMR.³ On the experimental point of view, two groups claimed the presence of an FeO intermixing layer,⁴ whereas we have never observed it yet.⁵ The best way to solve this problem is to perform direct measurements such as x-ray

photoemission spectroscopy (XPS) or/and absorption measurements on a unique Fe atomic layer in contact with MgO. This is one goal of this paper. Moreover, the presence of C contamination was underestimated and we will show that the oxygen adsorption on the Fe surface strongly depends on the presence of C. Furthermore, the effect of such C or O contamination at the interface on tunneling transport properties is studied by magnetotransport measurements on totally epitaxial Fe/MgO/Fe(100) MTJs. The samples were prepared by molecular-beam epitaxy. The growth, structure, and chemical aspects were characterized by scanning tunneling microscopy (STM), reflection high-energy electron diffraction (RHEED), Auger electron spectroscopy (AES), and x-ray spectroscopy (XPS). The Fe magnetic behavior was studied by x-ray magnetic circular dichroism (XMCD) at the Fe $L_{2,3}$ edges at room temperature.

II. EXPERIMENTAL RESULTS

A. Quality of the Fe(001) surface and the Fe/MgO(001) interface

The thin films are elaborated under a pressure less than 10^{-10} Torr. The substrates are monocrystalline MgO(001) outgassed up to 1070 K. A 50-nm-thick Fe layer is deposited at room temperature on these substrates using a Knudsen cell heated up at 1520 K. A postannealing at 920 K is thus nec-

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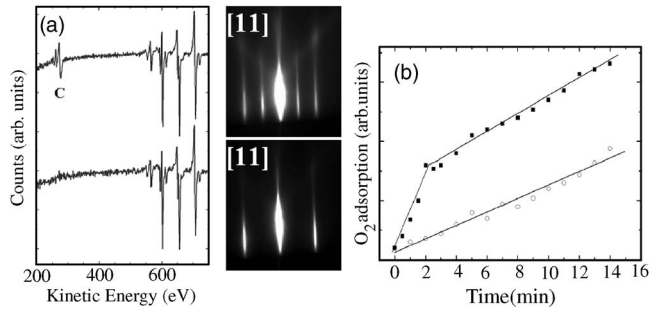


FIG. 1. (a) AES spectrum obtained on a C-contaminated Fe surface (top) and a clean Fe surface (bottom) and corresponding RHEED patterns along the [11] azimuth of the square lattice of Fe. Additional streaks for the C-contaminated Fe surface corresponding to a $C(2 \times 2)$ reconstruction are observed. (b) Arbitrary adsorbed O_2 quantity as a function of exposure time for a C-contaminated Fe surface (open circles) and for a clean Fe surface (full squares).

essary to smooth the surface. After this step, an Auger analysis shows the presence of C at the surface [Fig. 1(a)]. A typical $C(2 \times 2)$ reconstruction⁶ due to this contamination is observed by the RHEED patterns [Fig. 1(a)]. The signs of this reconstruction are additional streaks in the [11] azimuth. This C contamination comes from the MgO substrate itself. We observed that the outgassing step is not sufficient to desorb the entire C that can then diffuse and segregate to the Fe top surface. To eliminate this carbon, we deposit a 10-nm-thick interlayer of MgO at 1070 K prior to deposit Fe. The magnesium oxide is evaporated by means of electron bombardment at room temperature. By this way, a clean Fe surface is obtained as proved by Auger spectroscopy and RHEED patterns [Fig. 1(b)]. We are thus able to obtain two kinds of Fe surface called C-contaminated and clean Fe surfaces.

To test the chemical reactivity of these surfaces with oxygen, samples were exposed under a 10^{-8} Torr partial pressure of O_2 gas. The adsorption results are shown in Fig. 1(b). First, we clearly observe two different behaviors depending on the presence or absence of C. The O adsorption on the clean Fe surface is faster than on the C-contaminated one. The conclusion is that the surface without C is much more reactive to O than the unclean surface. Moreover, this reactivity is enhanced at the early stage of adsorption. The C could delay the oxidation reaction by site blocking. This high reactivity to O adsorption could be a disadvantage to obtain clean interface especially when the MgO deposition is performed. To test the quality of the Fe/MgO interface, we studied the electronic properties of 1 ML (monolayer) of Fe (grown on a Co buffer layer) in contact or not with the barrier. Details about samples and their preparation could be found in Ref. 7. The XPS measurements on the Fe monolayer are shown in Fig. 2 before [Fig. 2(b)] and after the MgO covering [Fig. 2(c)]. These spectra are compared to a reference bulk Fe spectrum obtained from a 50-nm-thick Fe [Fig. 2(a)]. These three spectra look very similar in shape and no shift in energy was detected. This means that no FeO formation occurs at the Fe/MgO interface according to our growth conditions.

In addition, the magnetic properties of the Fe monolayer at the interface with MgO (see Ref. 7 for details) were

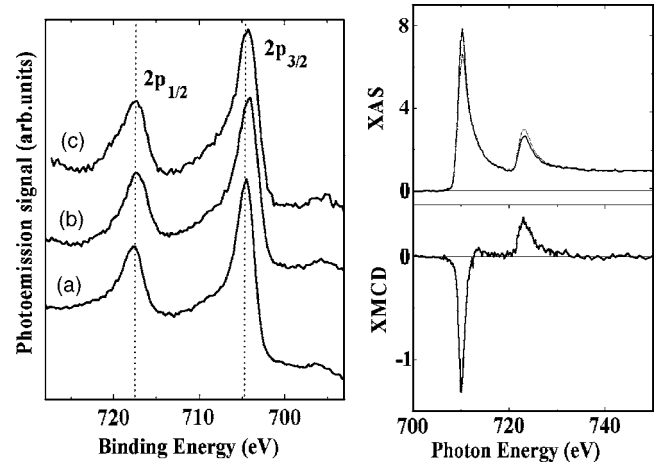


FIG. 2. Left: XPS spectra performed at room temperature on (a) a 50-nm-thick reference Fe layer, (b) bare Fe monolayer, and (c) same sample than (b) but covered with 2 ML of MgO. See text for details. Right: absorption and XMCD spectra performed at room temperature on Fe(1 ML)/MgO(1 nm) at the Fe $L_{2,3}$ edges. Note that all Fe surfaces of these samples are initially free of C or O contamination.

probed by XMCD at the Fe $L_{2,3}$ edges at room temperature. The experiments were performed at LURE on a Fe monolayer grown on a bcc Co film deposited on a V buffer layer. Again, the metallic shape of the Fe adsorption edge (Fig. 2) is a proof of the weak Fe/MgO hybridization. Indeed, the multiplet effect characteristic of 3d metals strongly hybridized with oxygen was not detected. A large dichroic signal is observed and the application of the sum rules leads to a magnetic moment of $3+/-0.3 \mu_B/\text{at}$. Finally, by using spin-resolved x-ray photoemission spectroscopy performed at ESRF on Fe covered by MgO, we have also shown for the first time that the Fe polarization was not killed by MgO.⁵ All these results confirm the theoretical predictions.^{2,8}

B. Transport measurements

The magnetotransport measurements were performed on UV photolithography patterned MTJs with the following stack: Fe(50 nm)/MgO(3 nm)/Fe(15 nm)/Co/capping layer. The typical junction size is $100 \times 100 \mu\text{m}^2$. Some typical results are shown in Fig. 3(a). TMRs as high as 160% are obtained at room temperature. The bias variation can be observed on a large range voltage ($+/-2$ V) and is highly symmetric meaning that the Fe/MgO and MgO/Fe interfaces are pretty similar. MTJs based on C-contaminated Fe surface was elaborated in order to make a comparison with the model interface based on initially clean Fe surfaces. Although high TMRs are also obtained (100%–150%), a very different bias variation is observed. The TMR(V) curve is highly asymmetric and an inversion of the TMR is observed.⁹ Finally, to study the influence of O contamination on the TMR, a monolayer of oxygen was inserted at the first interface leading to the following stack: Fe/O(1 ML)/MgO/Fe. This oxygen contamination leads to a drastic drop of the TMR. A maximum of 6% TMR is measured at room temperature.

These results from magnetotransport measurements show first that high TMR can be obtained from clean Fe

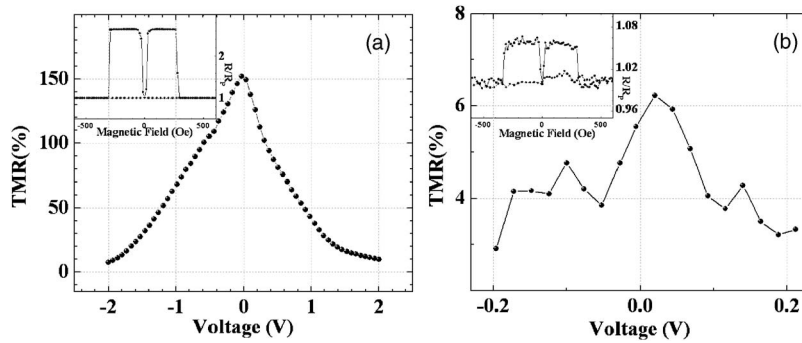


FIG. 3. Bias variation of the TMR. Inset: typical TMR vs field loop measured at room temperature and 10 mV for (a) a clean Fe/MgO interface and (b) a Fe/O/MgO interface. See text for details.

surfaces at room temperature. Second, the chemical nature of the contaminants at the Fe/MgO interface plays a crucial role in the tunneling process. An explanation is based on the fact that tunneling process in the Fe/MgO/Fe system is governed by spin filtering in the magnesium oxide barrier. Bloch states are more or less attenuated in the insulator as a function of their symmetry. The contaminant modifies the chemical bonding at the interface and modifies the spin injection efficiency in the barrier at the Fe/MgO interface.

III. DISCUSSION AND CONCLUSION

By controlling the C contamination at the Fe(001) surface, we have shown that the oxygen adsorption on this surface is strongly affected by the presence of C. Clean surfaces are observed to be highly reactive compared to the C-contaminated one, which could explain some spreading results in the literature. It should also be noted that careful attention must be paid during the preparation of the (001) Fe surface since the $C(2 \times 2)$ carbon surface structure is also a square lattice, and is often interpreted as the clean (001) square Fe lattice. Furthermore, despite this high reactivity to oxygen, we show that the Fe/MgO(001) interface is a model one from the chemical, structural, and magnetic points of view. In particular, no strong hybridization between Fe and O was detected at the Fe/MgO interface as shown by both XPS and absorption measurements performed on a unique atomic plane where all the Fe atoms are in contact with MgO. Large TMR is actually observed. Nevertheless, the most important result is that the influence of the C and O contaminations on

the TMR measurements is now clearly illustrated: the C contamination does not lead to a severe decrease of the TMR, on the contrary to oxygen contamination. To conclude, we have to point out that, if the first Fe/MgO interface is now well characterized, the situation is not so clear concerning the second MgO/Fe interface. Some recent works¹⁰ show that this interface seems not to be so different from the previous one. Finally, we performed a similar work on the bcc Co/MgO(001) interface,⁷ since the Co/MgO/Co(001) system is also expected to give large TMR. Like for the Fe/MgO interface, a weak hybridization between Co and MgO is observed, as well as an increase of the magnetic moment of Co in contact with MgO.

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