Spin-Dependent Quantum Interference from Epitaxial MgO Thin Films on Fe(001)

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(Received 19 May 2006; published 21 November 2006)

Spin-dependent electron reflection from MgO thin films grown on Fe(001) was measured using spin-polarized low energy electron microscopy. The electron reflectivity exhibits quantum interference from which two MgO energy bands with Δ_1 symmetry were determined in experiment. We found that a bulklike MgO energy gap is fully established for MgO film thicker than 3 atomic monolayers and that the electron reflectivity from the MgO/Fe interface exhibits a spin-dependent amplitude and a spin-independent phase change.

DOI: 10.1103/PhysRevLett.97.217205 PACS numbers: 75.70.Ak

Tunneling magnetoresistance (TMR) [1,2] in magnetic tunnel junctions (MTJs) has a high application potential in ''spintronics'' technology [3,4]. One key issue in MTJ is how to enhance the TMR value by improving the insulating layer property. In most MTJs, the insulating layer is made of amorphous aluminum oxide, and the best reported TMR value at room temperature is ~70% [5]. Recently, MgO insulating layers have been successfully grown onto Fe(100) single crystal substrates [6], and a room-temperature tunneling magnetoresistance of up to 220% has been achieved in Fe/MgO/Fe MTJs [7,8]. Moreover, a high spin polarization was also reported for current injection from a CoFe/MgO(100) tunnel injector into GaAs [9]. These discoveries highlight the crucial role of the electronic states of the insulating MgO in generating the large TMR effect. In fact, an extremely high TMR ratio (~1000%) was predicted by theory in Fe/MgO/Fe(100) MTJs [10,11]. Despite the importance of MgO as a spintronics material, not much is known about the electronic structure of MgO thin films, especially in terms of experimental data in the ultrathin regime. Theoretically, the MgO band structure has been calculated using different methods, and the results are not fully consistent with each other [12–17]. Experimentally, only the band gap and the surface states of MgO have been determined so far, using photoemission spectroscopy and electron energy loss spectroscopy [18,19]. Very little is known about the band structure of MgO ultrathin films, and this deficiency raises uncertainties about the basis of theoretical efforts to explain the TMR effect in Fe/MgO/Fe(100) MTJs. Obviously, it is very important to determine the electronic structure of MgO ultrathin films in experiment before a sound understanding of the extraordinary high TMR value in Fe/MgO/Fe MTJ can be developed.

In this Letter, we report an experimental study of the electronic band structure of MgO films grown on Fe(001). Using spin-polarized low energy electron microscopy (SPLEEM), we observed spin-dependent quantum interference from which we are able to determine the two energy bands of the MgO thin film with Δ_1 symmetry. In addition, our identical results from reflection asymmetry measurements on MgO films grown on clean Fe(001) and on films grown on $p(1 \times 1)$O-Fe(001) oxidized surfaces indicate that the Fe layer at MgO/Fe interface is in the oxidized state.

The experiments were performed in an ultrahigh vacuum system with a base pressure of 2×10^{-11} Torr. An Fe(001) single crystal disk was cleaned by cycles of Ar ion sputtering at 2 keV and annealing at ~900 K and confirmed by auger electron spectroscopy. MgO films were grown epitaxially on the Fe(001) substrate at room temperature using an *e*-beam evaporator, with the growth pressure remaining below 5×10^{-10} Torr. Recently, it was demonstrated that low energy electron microscopy can be applied to metallic thin films to retrieve the unoccupied energy bands [20–23]. Here we apply SPLEEM to characterize the growth and electronic structure of the insulating MgO films. A spin-polarized electron beam with ~30% polarization is directed to the sample surface at normal incidence, and the reflected specular beam is magnified by an electron-optical column to deliver the sample surface image. (A more detailed description of this instrument can be found in Ref. [24].) Prior to the MgO film growth, we first located one 180° domain wall of the Fe substrate and adjusted the spin polarization of the incoming electron beam to the direction of the Fe magnetization to achieve a maximum domain contrast of the SPLEEM image. In a typical experiment, we simultaneously deposited MgO onto the Fe substrate at a rate of ~0.04 ML (monolayer) per minute, recorded SPLEEM images at a rate of ~34 frames per minute, and simultaneously cycled the electron energy with 127 energy steps through a preset range. This type of experiment permits us to systematically measure the spin and energy dependence of the sample's electron reflectivity as a function of the MgO film thickness.

It was shown that MgO films grow pseudomorphically on Fe(100) in the ultrathin regime [25,26]. A detailed

scanning tunneling microscopy (STM) study [25] shows that the epitaxial growth of MgO on Fe(100) proceeds in a layer-by-layer manner which gives rise to a monolayer oscillation of the step density with the maximum step density occurring at half-integer layers. In our experiments, we find that the total electron reflection intensity (Fig. 1) has monolayer oscillations as a function of MgO film thickness, which are commonly attributed to the step-density oscillations associated with this type of layer-by-layer growth. Moreover, the oscillatory step density affects physical properties of the films in an oscillatory fashion. By measuring the electron mirror reflection energy [27], we find that the work function of the films oscillates with MgO thickness. This observation is consistent with previous results [28,29], which have shown that the work function of a film varies with local surface morphology, especially at the step edges. Our result (Fig. 1) shows that the work function drops rapidly during the initial growth (1–2 ML) and then exhibits a clear monolayer oscillation with peaks occurring at half-integer layers. The initial drop is due to the overall work function change from ~4.3 eV of Fe [30] to ~3.2 eV of MgO. The monolayer oscillation reflects the step-density oscillation during the epitaxial growth and the fact that the work function is slightly higher at the step edges than at the flat surface.

To obtain information of the electronic structure of the MgO film, we used unpolarized electrons to measure the electron reflectivity as a function of the electron energy and the MgO film thickness [Fig. 2(a)]. In the studied energy range (~4–23 eV), the reflectivity is strongly enhanced in the regions near $E < 4$ eV, 10 eV $< E <$ 13 eV, and $E >$ 21 eV for MgO films thicker than ~2–3 ML. This indicates the existence of energy gaps in these energy ranges. The monolayer reflectivity oscillation in these energy gaps is due to the surface morphology oscillation caused by the layer-by-layer growth. The existence of the energy gaps and their independence of the MgO thickness above ~2–3 ML MgO indicate that the MgO films on Fe(001) develop bulklike energy gaps above ~2–3 ML thickness. This result is consistent with an STM study of the MgO/Ag(001) system, which shows that the MgO film develops an insulating energy gap at the Fermi level above 3 ML thickness [18]. Outside the energy gaps, where electrons can occupy states within the MgO energy bands (4–10 and 13–21 eV), we observed clear quantum well (QW) interference effects. The electron reflectivity reaches a maximum at QW resonance condition due to the Febry-Pérot interference effect between the electrons reflected at the MgO/vacuum surface and the MgO/Fe interface [22]. To highlight the QW interference, we normalized the spectra of MgO thin films by the bulk MgO spectrum (obtained from a thick MgO film) and display the result in Fig. 2(b). The QW states exhibit discrete peaks only at integer layers of the MgO thickness. Such behavior exists only in atomically flat thin films [23,31], confirming again the high quality of our MgO films on Fe(001). Since the observed QW states are in the unoccupied band above the vacuum level, it leaves open whether the quantization of electronic states could also exist in the occupied band of the MgO insulating layer, similar to the QW states observed by photoemission in insulating noble-gas layers at cryogenic temperatures [32].

The QW states are usually described by the so-called phase accumulation model which gives the quantization condition of [33]

$$2k(E)d_{\mathrm{MgO}} + \phi_B(E) + \phi_C(E) = 2n\pi, \qquad n = \text{integer}. \tag{1}$$

Here d_{MgO} is the MgO film thickness, $k(E)$ is the electron wave vector, and $\phi_B(E)$ and $\phi_C(E)$ are the phase gains of the electron upon reflection at the two surfaces of the MgO film, respectively. Since the phase value depends on

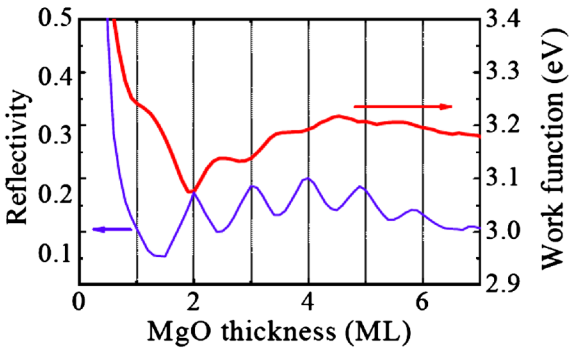

FIG. 1 (color online). Thickness dependence of the electron reflectivity (at 4 eV) and work function of the MgO thin film grown on Fe(001).

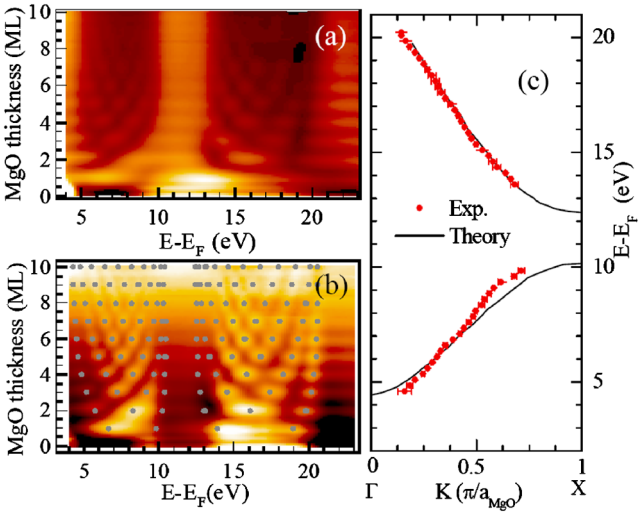

FIG. 2 (color online). (a) Electron reflection intensity versus electron energy and MgO film thickness. (b) Reflectivity normalized by the spectrum of bulk MgO. The dots mark quantum well resonance positions as described in the text. (c) MgO unoccupied energy bands with D_1 symmetry. The solid lines are from band structure calculation [16]. The calculated energy bands have been offset to best fit the experimental data.

the electron energy only, the electron wave vector at a given energy depends only on the thickness periodicity $[\Lambda(E)]$ of the QW state oscillations with $k(E) = \pi/\Lambda(E)$. Therefore, the energy band dispersion of the MgO film can be derived entirely from the QW experimental data without the need of any models for the phase value [21]. Figure 2(c) shows the two MgO energy bands determined from the QW oscillations in the energy ranges of 4–10 and 13–21 eV. As a comparison, we also show [solid lines in Fig. 2(c)] the corresponding bulk MgO energy bands (Δ_1 symmetry) from an *ab initio* calculation [16]. While there is an overall agreement, the slight difference between the theoretical and the experimental results, especially for the lower energy band, may come either from the fact that the energy band of a MgO thin film is slightly different from the bulk band or from the fact that the band structure calculation is not sufficiently accurate. Using the experimentally determined energy bands, we fit the QW states positions in the *E-d* plane [Fig. 2(a)] by adopting a linear dependence of the total phase shift on the energy [34]. The calculated QW positions are depicted as dots in Fig. 2(b). The excellent agreement between the experiment and the fitting indicates that the MgO electronic structure in MgO/Fe(100) is well stabilized above ~3 ML MgO thickness, and the TMR reduction in Fe/MgO/Fe junction below ~20 Å MgO is not due to thickness-dependent electronic structure changes of the MgO film but rather due to other possible mechanisms [10,35].

Figure 3(a) shows the spin asymmetry of the reflectivity as a function of the electron energy at different MgO film thicknesses. Here the spin asymmetry is defined as $A \equiv (R_P - R_{AP})/(R_P + R_{AP})$, where R_P (R_{AP}) is the reflectivity with the electron spin parallel (antiparallel) to the Fe magnetization direction. We found a clear nonvanishing spin asymmetry in electron reflection from the MgO film up to at least 10 ML thickness. Moreover, the spin asymmetry oscillates not only with the electron energy but also with the MgO thickness [Fig. 3(b)]. However, for electron reflection within the MgO energy gap, the spin asymmetry vanishes above 3–4 ML MgO due to the very short attenuation length of the evanescent gap states. The spin asymmetry exhibits a clear interference effect in the energy range of the MgO energy bands. Noting that the reflectivity oscillation is a result of interference between electron waves reflected from the vacuum/MgO and MgO/Fe interfaces, the nonvanishing spin asymmetry shows that the electron reflectivity at the MgO/Fe interface is spin-dependent. In another words, the MgO electronic state with Δ_1 symmetry is strongly coupled to the electronic state of the Fe substrate at the MgO/Fe interface. In principle, the spin asymmetry oscillation could come either from the amplitude or from the phase difference of the spin-up and spin-down reflectivity oscillations. To distinguish these two mechanisms, we plot R_P and R_{AP} versus the MgO thickness in Fig. 4. Both R_P and R_{AP} oscillate with MgO thickness due to the interference effect, but R_P and R_{AP} have exactly the same peak positions with only their amplitudes being different. Therefore, the spin asymmetry of the electron reflectivity originates from the amplitude, not the phase of the reflectivity R_P and R_{AP}; i.e., the reflectivity at the MgO/Fe interface has a spin-dependent amplitude but a spin-independent phase. This result is different from the result of the metallic Cu/Co system, where both the amplitude and the phase of the reflectivity are spin-dependent [20,22]. This observation may be helpful for further understanding of the spin-dependent transport across the MgO/Fe interface.

It was reported that the Fe/MgO interface is actually composed of an FeO layer [6,36], which could have a profound effect on the electrical conductivity in the

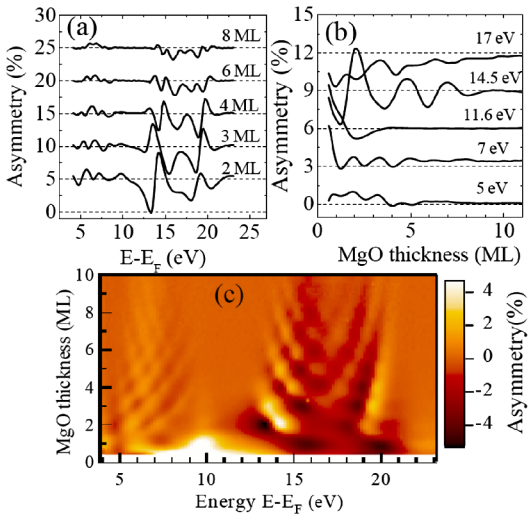

FIG. 3 (color online). (a) Electron reflection asymmetry as a function of energy for MgO film for several thickness values, as labeled on the right. (b) Electron reflection asymmetry as a function of MgO thickness for several different values of the electron energy, as labeled on the right. The horizontal dashed lines represent zero spin asymmetry for each spectrum. (c) Composite reflection asymmetry versus electron energy and MgO thickness.

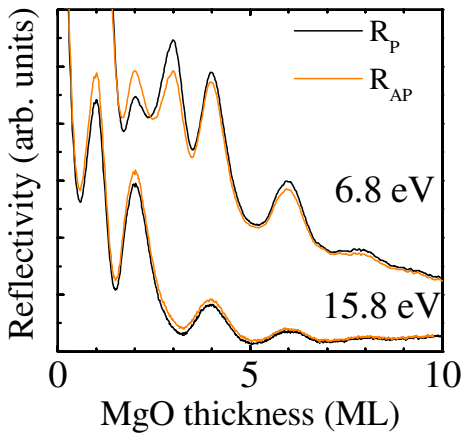

FIG. 4 (color online). Spin-dependent electron reflectivity versus MgO thickness at different electron energies.

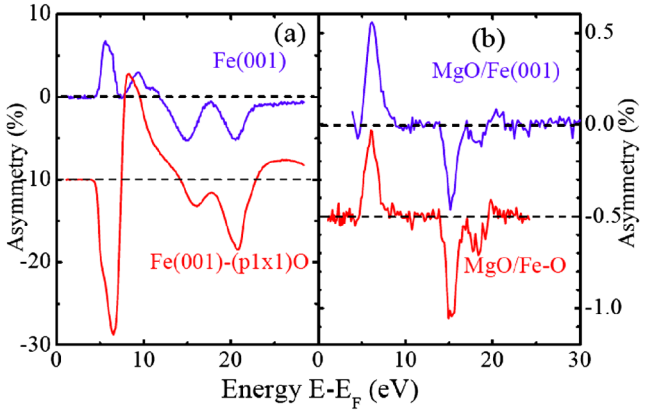

FIG. 5 (color online). Asymmetry of the electron reflectivity as a function of the electron energy (a) from a clean Fe(001) surface and a Fe(001)-($p1 \times 1$)O oxidized surface and (b) from a 14 ML MgO/Fe(001) and 14 ML MgO/Fe(100)-$p(1 \times 1)$O surface. The horizontal dashed lines represent zero spin asymmetry for each spectrum.

Fe/MgO/Fe system [37]. To further explore the electron reflectivity at the MgO/Fe interfaces, we studied the electron reflectivity from oxidized Fe(001) surfaces. We prepared well ordered stable Fe(001)-$p(1 \times 1)$O surfaces [38] by exposing the Fe(001) surface to 10 L O_2 at room temperature and then flashing the substrate at 900 K for a few seconds [30]. Compared to the clean Fe(001), the Fe(001)-$p(1 \times 1)$O surface shows a different spin asymmetry in reflection [Fig. 5(a)], especially near $\sim$6 eV electron energy, which is consistent with previously reported results [30]. After growing 14 ML MgO on the Fe(001)-$p(1 \times 1)$O surface, we measured the spin asymmetry of the electron reflectivity and compared the result with that of 14 ML MgO grown on clean Fe(001). We found identical reflection asymmetry for MgO films grown on clean Fe(001) and on a Fe(001)-$p(1 \times 1)$O surface [Fig. 5(b)], supporting our conclusion that the Fe at the MgO/Fe(001) interface is in the oxidized state [6,36].

In summary, we studied spin-dependent electron reflectivity from MgO thin films grown on Fe(001) using SPLEEM. Quantum interference states were observed from which we derived the two MgO unoccupied energy bands with Δ_1 symmetry. We found that the bulklike MgO electronic structure is fully developed for MgO film thicker than 3 ML. Moreover, the two MgO energy bands with Δ_1 symmetry are coupled to the Fe substrate electronic states to result in a spin-dependent electron reflectivity from the MgO films. Our results show that this spin dependence arises from a spin-dependent reflection amplitude alone, while the phase change at the MgO/Fe interface is spin-independent. We also confirmed that the Fe at the MgO/Fe interface is in the oxidized state.

This work was supported by National Science Foundation No. DMR-0405259, U.S. Department of Energy No. DE-AC03-76SF00098, DARPA-ONR Grant No. 00014-96-1-1207, National Natural Science Foundation of China and Shanghai Science and Technology Committee, ICQS of Chinese Academy of Science, and SRF for ROCS, SEM of China.

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