Tunneling magnetoresistance in epitaxial discontinuous Fe/MgO multilayers

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Epitaxial discontinuous Fe/MgO multilayers have been grown by pulsed laser deposition on MgO(001) single-crystal substrates. The multilayers with 0.6 nm nominal Fe layers thickness are superparamagnetic and demonstrate tunneling magnetoresistance (TMR) in the current-in-plane configuration. Increasing deposition temperature causes an improvement in crystal quality and is accompanied by higher TMR ratios. The maximum value (9.2% TMR at room temperature and 18 kOe magnetic field) trebles that of polycrystalline samples deposited simultaneously on glass substrates. A model formula for TMR ratio that includes both spin-dependent tunneling and spin-filtering effect is proposed to explain these results. © 2011 American Institute of Physics. [doi:10.1063/1.3569149]

Discontinuous metal-insulator multilayers (DMIMs) are cermet structures prepared by repeated alternate deposition of a continuous insulating and a discontinuous metal layer composed of nanometer-sized islands. The size and shape of the nanoparticles can be controlled by the nominal thickness of the metallic layer (typically of the order of 1 nm) and deposition parameters. Ferromagnetic metal-based DMIMs show moderate room-temperature (RT) tunneling magnetoresistance (TMR) that originates from spin-dependent tunneling between adjacent metallic granules.^{1–5} Significant low-field sensitivity in the current-in-plane (CIP) configuration and relative ease of preparation make DMIMs promising materials for sensors and read heads.³ Although DMIMs were actively studied during the past decade, only structures with amorphous insulator layers have been prepared so far by different groups (see for instance Ref. 5 and references therein). No studies on the influence of the substrate nature, deposition temperature, and degree of crystallinity of the insulator layer on transport properties of DMIMs have been reported previously.

In planar magnetic tunnel junctions (MTJs) a substantial increase in TMR ratios appeared when single-crystal MgO replaced amorphous alumina tunnel barriers.⁶ This is due to the crucial role of the electron band symmetry of the electrodes and the barrier in epitaxial Fe/MgO/Fe MTJs.^{6,7} Recently,^{8–10} we studied Fe/MgO DMIMs deposited on glass substrates by pulsed laser deposition (PLD). In these materials the TMR ratio is defined as $TMR = 100 \times [R(H) - R(H=0)]/R(H=0)$, R being the electrical resistance and H the

magnetic field. For Fe layers with 0.61 nm nominal thickness, TMR ratios $\approx -3\%$ at RT were achieved in 18 kOe magnetic field. In this case, the MgO spacers grew in polycrystalline state with a tendency to form large columnar grains through the Fe layers. This suggests that increasing the degree of texturing of Fe/MgO DMIMs could lead to an enhancement of TMR.

In this letter, we report on the preparation, structural characterization, and CIP TMR of epitaxial Fe/MgO DMIMs deposited by PLD on single-crystal MgO(001) substrates. The effect of elevated substrate temperature during deposition (T_S) on the microstructure and transport properties is studied. Comparison with films prepared in identical technological conditions on amorphous substrates is made.

The deposition system for the preparation of $MgO(3\text{ nm})/[Fe(0.6\text{ nm})/MgO(3\text{ nm})]_{10}$ DMIMs was described in Ref. 8. To induce textured growth of the multilayers we used single-crystal MgO(001) substrates $5 \times 5\text{ mm}^2$ in size (Crystal GmbH). For comparison, Corning No. 0211 glass substrates (CG) of the same size were placed aside on the sample holder, which was rotated during deposition to ensure symmetry relative to the plume. The effect of the growth temperature was studied for four values of T_S : 20, 120, 180, and 250 °C.

CIP electrical transport measurements were carried out at RT using the four-probe technique and experimental setup described elsewhere.^{9,10} RT TMR values for the films deposited on CG are $\sim -3.5\%$ at 18 kOe and show negligible dependence on T_S [Fig. 1(a)]. On the contrary, DMIMs deposited on MgO(001) present enhanced TMR with increased T_S , reaching the absolute value 9.2% (for $T_S = 250\text{ °C}$) [Fig.

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FIG. 1. (Color online) RT TMR vs applied magnetic field (H) measured for Fe (0.6 nm)/MgO (3 nm) DMIMs deposited on (a) CG and (b) MgO(001) substrates at the indicated values of T_S .

1(b)]. Structural and magnetic characterization of the films reveals the cause of this increase.

Cross-section high-resolution transmission electron microscopy (HRTEM) investigations of the samples grown on MgO(001) and CG were carried out in a FEI Tecnai F30 transmission electron microscope operated at 300 kV. All our samples deposited on CG are polycrystalline, as is the case for those reported earlier in Ref. 8. On the contrary, the four samples on MgO(001) show epitaxial growth of MgO alternating with Fe layers and with structural coherency along the whole thickness of the multilayer, even for $T_S = 20$ °C (Fig. 2). The crystal coherence shown in this HRTEM cross-section has never been observed before in DMIMs, due to the polycrystalline or amorphous nature of both the metal granules and insulating matrix. Dislocations and other structural defects caused by lattice mismatch between MgO and Fe (Ref. 6) can be seen at high magnification. Although the crystal orientation of Fe cannot be elucidated from our observations, three-dimensional epitaxy between Fe islands and MgO matrix would be possible through

FIG. 2. HRTEM cross-section image of the Fe (0.6 nm)/MgO (3 nm) DMIM deposited at 20 °C on MgO(001).

FIG. 3. (Color online) (a) $\theta/2\theta$ scans measured in the Fe (0.6 nm)/MgO (3 nm) DMIMs deposited on MgO(001) at the values of T_S indicated. (b) Average interplanar spacing d obtained from Bragg's law at the film diffraction maxima in (a) (circles), and full width at half-maximum of the rocking curves taken at the same maxima (squares). The continuous lines are guides to the eye.

the epitaxial relationships reported in coevaporated granular Fe/MgO cermets:¹¹ $(001)[110]_{\text{Fe}} \parallel (001)[100]_{\text{MgO}}$ and $(011)[100]_{\text{Fe}} \parallel (001)[100]_{\text{MgO}}$.

TEM micrographs provide only some clues about the discontinuous nature of the Fe layers (Fig. 2), due to the fact that TEM specimens' thickness (a few tens of nanometers) is much larger than typical Fe granule size. This causes superimposition of the granules over the specimen thickness and gives the impression of a continuous Fe layer in many areas. Fe is well known to follow three-dimensional (Volmer–Weber) nucleation and growth on MgO(001) (Ref. 12), so that only for sufficient thickness, typically above 1 nm, continuous films appear as a result of islands coalescence.^{12–14}

The discontinuous nature of the Fe layers was confirmed by zero-field cooled (ZFC) and field cooled (FC) magnetic susceptibility measurements. They were carried out using a Quantum Design MPMS magnetometer in $H = 50$ Oe and in the temperature range 5–300 K. A maximum associated with progressive unblocking of small superparamagnetic granules is present on ZFC curves for all the investigated samples. The position of the maximum is shifted toward lower temperature and its width decreases with increasing T_S . The bifurcation temperature, T_B (the one below which the ZFC and FC curves split) for samples deposited on MgO(001) monotonically decreases from 180 to 45 K as T_S increases from 20 to 250 °C. Very similar variation in T_B from 200 K to 50 K for the same deposition conditions was found for the samples on CG substrates. As the same amount of Fe has been deposited in all the samples, reduction in T_B points that increasing T_S leads to higher nucleation density,¹⁵ average Fe granule size decrease, and narrowing of the particle sizes distribution.⁸ The detailed analysis of magnetic properties and Fe particles size and shape evolution with T_S will be the subject of a forthcoming publication.

The evolution of the crystal quality of the multilayers deposited on MgO(001) with T_S was studied by x-ray diffraction using a Bruker D8 high-resolution diffractometer with $\text{Cu K}\alpha_1$ radiation. Symmetrical $\theta/2\theta$ scans in the 2θ range 41° – 44° [Fig. 3(a)] show the 002 reflection from the MgO substrate at $2\theta \approx 42.9^\circ$ and a strong reflection around $2\theta \approx 42^\circ$ which is the result of the coherent growth of continuous epitaxial MgO with varying degrees of lattice distort-

tion for different values of T_S . In Fig. 3(b), we show the average interplanar spacing in the out-of-plane direction obtained through Bragg's law, $d = \lambda / 2 \times \sin \theta$ (λ is the x-rays wavelength), which shows a tendency to approach the bulk MgO value $d = 2.106 \text{ \AA}$ upon increasing T_S . This might be related to the decrease in the Fe particles size deduced from magnetic measurements. The width of the rocking curves taken at the maximum of the film reflection decreases with increasing T_S [Fig. 3(b)], indicating improvement of the crystal quality. The coherence length determined through Scherrer equation agrees with the multilayer thickness deduced from x-ray reflectivity for any value of T_S , thus, confirming that the multilayers show crystal coherence along their whole thickness.

Experimental data allow us to conclude that increasing T_S causes both an enhanced degree of (001) texturing of MgO and a decrease in the average Fe granule size. At the same time, an increase in TMR value is observed. It has been previously shown that MgO (001)-oriented barriers produce electron band symmetry filtering.^{6,7} Thus, electrons from Δ_1 band from Fe(001) electrodes tunnel more efficiently through MgO(001) than the Δ_2 and Δ_5 ones. The absence of Δ_1 in the spin-down subband of Fe(001) causes the large TMR ratios observed in epitaxial Fe(001)/MgO(001)/Fe(001) MTJs. It is feasible that the increase in the degree of (001) texturing in our Fe/MgO DMIMs favors this type of band symmetry filtering effect and leads to TMR enhancement. On the contrary, the samples deposited on CG grow without preferential orientation and show a negligible variation in TMR with T_S . These effects can be modeled through

$$\text{TMR} = \frac{P^2 + P\varphi}{1 + P^2 + 2P\varphi} (1 - \cos \psi), \quad (1)$$

an extension of the known Inoue–Maekawa formula¹⁶ for TMR, with spin polarization P and average angle ψ between adjacent granules moments, enhanced by a nonzero spin-filtering factor $\varphi = (t_{\uparrow} - t_{\downarrow}) / (t_{\uparrow} + t_{\downarrow})$, where t_{σ} is the full tunnel rate in σ spin channel. This formula explains the lower TMR for samples grown at 20°C on MgO substrate – φ is still too small to give advantage and ψ is lower than for samples grown on glass due to the similar orientation of crystalline anisotropy axis in neighboring granules because of the epitaxial growth. For $P = 0.3$ (Ref. 9) the change in φ from 0 to 1 will increase TMR by a factor of 3, in good agreement with our results on MgO(001) substrates.

In conclusion, epitaxial Fe/MgO DMIMs have been prepared. Elevated deposition temperature causes the improve-

ment in crystal quality and is accompanied by an increase in TMR. The absolute value of TMR ratio 9.2% at RT in 18 kOe has been achieved, and higher values may be expected by optimizing deposition conditions and measurement geometry. This result demonstrates an alternative route to prepare DMIMs with improved magnetotransport properties.

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