Reduction of spin polarization by incoherent tunneling in Co$_2$FeAl/MgO/CoFe magnetic tunnel junctions with thick MgO barriers

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We investigate incoherent spin polarized tunnel transport regime in single crystal Co$_2$FeAl/MgO/Co$_{50}$Fe$_{50}$ magnetic tunnel junctions (MTJ) with 4.2 nm thick MgO barriers grown by combined Molecular Beam Epitaxy and Sputtering deposition techniques. Tunnel transport spectroscopy experiments at variable temperature correlated with High Resolution Transmission Electron Microscopy phase analysis demonstrate the effect of the high density misfit dislocations within the thick insulator on the longitudinal and lateral coherence of the propagating Bloch functions. These dislocations are extended defects within the barrier which destroy the symmetry and $k$ conservation of the electron wave function across the stack affecting drastically the TMR ratio of the MTJs. The reduction of defects density within the barrier in fully sputtered MTJ devices leads to one order of magnitude increase of the TMR.

I. INTRODUCTION

The performance of spintronic devices based on magnetic tunnel junctions (MTJs) is mainly dependent on their tunnel magnetoresistive (TMR) response amplitude. In epitaxial magnetic tunnel junctions, the TMR is a consequence of several factors: the spin polarization of the ferromagnetic electrodes, the symmetry filtering properties of the electrodes and the symmetry dependent attenuation rate of the evanescent wave function in the tunnel barrier. Consequently, two alternatives are employed in order to increase the TMR response of a MTJ. One is to use half-metallic ferromagnets (HMFs) as magnetic electrodes in MTJ. Since they have an energy gap around the Fermi level $E_F$ in the minority spin band, theoretically, they are expected to provide 100% spin polarization. The other alternative is to use $\Delta_1$ band metals [1] and alloys, [2,3] which in conjunction with an epitaxial (100)MgO barrier behave like half metals in the coherent tunneling regime [4,5]. Among the HMFs, a special class is represented by the full-Heusler alloys. Theoretical predictions [6,7] indicate that the Co-based full-Heusler alloys should behave like half-metals even at room temperature. Currently, one of the most studied full-Heusler alloy is Co$_2$FeAl (CFA). It was demonstrated to provide giant tunneling magnetoresistance (GTMR) in MgO based MTJs due to the mixed effect of large spin polarization and $\Delta_1$ band symmetry filtering [8,11]. Moreover, this material is of special importance since it was shown to have a low Gilbert damping [12,13], essential for magnetization reversal by spin-torque using low current densities and for building high efficiency spin torque oscillators.

In this paper we address a special tunneling transport regime in CFA Heusler based MTJs, corresponding to thick MgO barriers. By detailed structural analysis correlated with tunneling transport we demonstrate that the lateral and longitudinal coherence of the propagating wave function is reduced. By symmetry breaking due to extended spatial defects related to oblique dislocations, the wave vector $k$ is not conserved and incoherent tunnel transport channels become dominating.

II. RESULTS AND DISCUSSIONS

MTJs with the structure MgO(001)//Cr(20nm)/CFA(40nm)/MgO(4.2nm)/CoFe(10nm)/Co(40nm)/Au(10nm), have been grown by combining two deposition techniques: sputtering and Molecular Beam Epitaxy (MBE). The Cr buffer layer and bottom CFA electrode have been grown in a magnetron sputtering system, as described elsewhere [13,14]. Afterwards, the MgO//Cr/CFA stack has been transferred in an MBE chamber equipped with reflection high energy electron diffraction (RHEED) system, as described elsewhere [13,14]. Afterwards, the MgO//Cr/CFA stack has been transferred in an MBE chamber equipped with reflection high energy electron diffraction (RHEED) analysis. In Fig. 1a, we illustrate the RHEED pattern of the as deposited CFA film.

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FIG. 1. RHEED patterns of (a) as deposited CFA film and (b) annealed at 600°C; (c) RHEED intensity oscillations for a MgO film grown on CFA and Cr; RHEED patterns of the MgO film after growth on (d) CFA and (e) Cr.

The film is crystalline but rough within the RHEED coherence scale-length, the diffusive background indicating chemical disorder present in the as-deposited sample. After annealing at 600°C for 20 minutes, the CFA layer is flattened, the reduction of the diffusive background being correlated with the improvement of the chemical ordering of the system from A2 towards the B2 phase [14]. On top of this CFA layer cooled down to 120°C, the MgO barrier has been grown by electron beam evaporation within the MBE chamber. During growth, the pressure in the deposition chamber increased from $5 \times 10^{-11}$ Torr to $10^{-8}$ Torr due to the enhancement of oxygen partial pressure as confirmed by Quadrupole Mass Spectrometry.

The growth of MgO on CFA has been carefully monitored by RHEED. Analysis of intensity oscillations (Fig. 1c) demonstrates a 3D growth mode of MgO on CFA. The oscillations are damped rapidly, in contrast with the growth of MgO on (001) Cr (Fig. 1c) where a clear layer-by-layer growth is asserted by the RHEED intensity oscillations with a low attenuation. Moreover, the 3D growth of MgO on CFA is confirmed by the final RHEED pattern of a 4.2 nm thick MgO barrier (Fig. 1d). We observe that the MgO exhibits a poorly (001) crystallized structure mixed with a polycrystalline phase and a rough surface within the RHEED coherence scale, probably related to the complex chemical structure of CFA surface in B2 phase (random distribution of Fe and Al atoms within their atomic sites). In contrast, similar RHEED pattern recorded for a 12ML MgO grown on Cr (corresponding to the RHEED oscillations from Fig. 1c), show that in this case the barrier grown on Cr is flat and with an almost perfect (001) orientation.

In our study, we used the standard growth of MgO on Cr to calibrate the deposition rate of MgO. Due to the 3D growth of MgO on CFA, the accurate RHEED intensity oscillations technique, typically used for monitoring in-situ the thickness of the barrier when growing epitaxial MTJs [15], cannot be applied (Fig. 1c). Therefore, our strategy consists to use and extrapolate the calibration obtained from the growth of MgO on Cr (in terms of MgO mass variation recorded by a quartz balance correlated with RHEED intensity oscillations) in the case of the MgO growth of CFA. In this way, we intended to have about 12ML thick MgO barrier on CFA using the same quartz balance indication as the one corresponding to 12 ML of MgO on Cr. However, further detailed analysis (TEM and transport) performed in CFA/MgO MTJs demonstrate that the error concerning the estimated barrier thickness was very large: the final MgO barrier on CFA was found to be about 20 ML thick with respect to 12ML targeted. This could be probably explained by the growth mode of MgO on CFA, which is not layer-by-layer, as in the case of MgO growth on Cr, combined with different adhesion coefficient of MgO on CFA and quartz balance monitor intrinsic errors, which are always larger than 10%. We underline the fact that during the MBE growth when the RHEED intensity oscillations technique cannot be applied (i.e. the growth is not layer-by-layer) the in-situ estimation of the thickness becomes problematic especially for the tunnel barrier for which the thickness has to be drastically controlled. Thickness calibrations from ex-situ measurements (i.e. X-Ray reflectometry) are not useful since deposition rate can change from one growth to the next one. Having in view the exponential dependence of the tunneling current with the barrier thickness, this uncertainty of the barrier thickness is of major importance in magnetic tunnel junctions. We will show later in our study that the under-estimated errors on the MgO barrier thickness lead to major transport and spin filtering problems.

After deposition of the barrier, the top CoFe magnetic electrode of the junction has been grown by MBE at room temperature (RT). As expected from the poor crystal quality of the MgO barrier, the as-deposited CoFe layer consists of a (001) crystallized structure mixed with a polycrystalline phase (Fig. 2a). However, an in-situ annealing of the CFA/MgO/CoFe stack at 450°C promotes the layers crystallization improving also the surface flatness (Fig. 2b), as confirmed also by the X-ray 2theta-theta diffraction pattern (Fig. 2c).
Further structural analysis have been performed by high resolution electron transmission microscopy (HRTEM) on cross-sectional specimen. The HRTEM images demonstrate a good crystalline quality and relative flat interfaces (Fig. 3a). This confirms that the annealing stage of the upper CoFe electrode triggers the crystallization of the MgO tunnel barrier and improves the quality of the interfaces. However, accurate analysis of the interface roughness by TEM is delicate because it integrates the roughness profile over the analyzed section width. Measurements of the atomic planes spacing show that the MgO barrier is totally relaxed, d(002) being equal to d(200) and with a value of 0.21 nm, which is the value expected for a bulk MgO crystal. The lattice mismatch between CFA and MgO is about 3.8%, which gives considerable strain that relaxes through the formation of misfit dislocations. Dislocations and strain fields can be visualized in HRTEM by performing the so-called ”Geometrical Phase Analysis” (GPA) procedure [16] (Fig. 3b,c,d). A periodic array of additional lattice planes perpendicular to the interfaces corresponding to misfit dislocations, can be visualized in the electrodes (Fig. 3b). The dislocations are perpendicular to the image plane and have a periodicity of approximately 4.4 nm, and are correlated from one side of the barrier to the other. This becomes even clearer from the Fig. 3c which shows the missing planes within the insulating barrier. One can see that the dislocations from the CFA electrode continue throughout the barrier and to the upper electrode. Inside the barrier, among the misfit dislocations that are relatively perpendicular to the interfaces, one can observe dislocations that impinge on the interface at much smaller angles and continue over tens of nanometers. Because they destroy the local symmetry of the crystal and act as scattering centers, the dislocations have a major impact on the spin dependent tunneling. Figure 3d shows the local deformation field $\varepsilon_{yy} = du_{yy}/dy$ in the direction parallel to the interface. The lattice deformations are concentrated mostly at the interfaces and inside the barrier and are correlated with the misfit dislocations. From the HRTEM analysis two important conclusions can be drawn. First, because it is not possible to observe RHEED intensity oscillations during the growth of MgO on CFA, the accurate control of the barrier thickness is a very delicate procedure, the thickness of the insulator being almost double than expected. The second is that, even though the crystalline quality of the barrier increases after the annealing of the upper CoFe electrode, it contains a large density of defects, like lattice deformations and dislocations that, as we will demonstrate in this paper, will affect the spin dependent tunneling process.

To operate magnetically the tunnel junction, a hard-soft architecture is required. In our MTJ stack, the magnetic hardening of the CoFe layer is obtained by direct exchange coupling with an additional 40nm thick Co film [17]. This Co film grows epitaxially on CoFe in a hexagonal phase with the c axis parallel to the film plane. The
MTJ stack has been magnetically characterized by vibrating sample magnetometry using the vector field option. Detailed information about the magnetic properties of the CFA films and their tuning by thermal annealing can be found in our previous work [14].

In order to perform magneto-transport experiments our MTJ stacks have been patterned by UV lithography and ion beam etching in square MTJ devices with lateral size from 10 to 50 µm. In Fig. 4a we illustrate the tunnel magnetoresistance (TMR) curves for the CFA/MgO/CoFe MTJ measured both at low (14K) and at room temperature (300K). The first remark is the low TMR ratio at room temperature (2.8%), in contrast to standard results obtained in similar systems reported in literature (larger than 300% ratios at RT in CFA/MgO single crystal MTJs, 166% at RT in textured CFA/MgO MTJs [8–11]). This TMR increases to 20% when the temperature is lowered. In order to understand the origin of this low TMR ratio, unusual for single crystal MTJ devices, we performed several sets of additional experiments in order to correlate the magneto-transport characteristics with the structural analysis.

In Fig. 4b we illustrate the variation of the MTJ resistance in the parallel (P) and antiparallel (AP) magnetization configurations as a function of temperature. In contrast to standard R_P(T) and R_AP(T) measured in single crystal MgO based MTJs, where R_P(T) has been found nearly constant [18, 19], here both R_P and R_AP have a strong and similar dependence with the temperature. The enhancement of the TMR at low temperatures is related to a relatively higher variation of R_AP with T. Even if this is a common feature of single crystal MTJs, the relative variation of R_AP recorded here is extremely large (800% with respect to less than 100% measured in standard Fe/MgO/Fe MTJs). Moreover, the variation of R_P is about 650% from 300 to 14K. These large variations indicate that the tunneling transport in our MTJs is dominated by non-coherent transport channels in both P and AP configuration, most probably related to defects within the tunnel barrier. The large measured R × A values of our MTJs (10^8 to 10^9 Ω x µ² range), are correlated with the large thickness of the MgO barrier. They are consistent with extrapolated R × A from values measured in single crystal Fe/MgO/Fe MTJs [1, 15].

In order to clearly show the dominating incoherent transport in our thick barrier CFA/MgO/CoFe MTJs, detailed structural investigations by HRTEM phase image contrast analysis have been performed. These results have been correlated with tunneling spectroscopy experiments (dI/dV curves) measured in patterned MTJ devices. The results (Fig 5a,b,g) were compared to those obtained in standard Fe/MgO/Fe MTJs (Fig. 5c,d,h) where we have previously demonstrated that tunneling coherent spin and symmetry dependent tunneling channels dominate the conductance [15] [17] [20]. Fig. 5b illustrates the phase shift image corresponding to the real space image of Fig. 5a showing the presence of the misfit dislocations inside the thick MgO barrier grown on (100) CFA. The density of dislocations is significantly larger than the one corresponding to the MgO barrier grown on Fe (Fig. 5d). Consequently, as sketched in Fig. 5e and Fig. 5f, no coherent tunneling channel with conservation of k_∥ = 0 (denoted here by CH1) can be defined in the MgO barriers grown on CFA, whereas in the case of the thinner MgO barrier grown on Fe coherent tunneling channel are present. Therefore, the tunneling transport in the case of the thick MgO barrier will be dominated by incoherent tunneling channels (denoted CH2) where the dislocation related defects will first quench the symmetry filtering and secondly will induce, by elastic scattering on the perturbations in the potential profile seen by the propagating electron, violation in k conservation for the tunneling electrons. In these incoherent transport channels, the temperature influence on the tunneling conductivity can be significantly large and explain the experimental R(T) variation measured in our MTJs. From the tunneling transport point of view (tunneling spectroscopy experiments), the signature of the incoherent transport channels is given by the parabolic dynamic.
FIG. 5. (a) HRTEM image of the CFA/MgO/CoFe trilayer and (b) corresponding phase image; (c) HRTEM image of the Fe/MgO/Fe trilayer and (d) corresponding phase image; (e) and (f) schematic representation of the misfit dislocations inside the MgO barrier as resulted from (b) and (d), the coherent (CH1) and incoherent (CH2) conduction channels are also schematized; (g) and (h) bias voltage dependence of differential tunneling conductance $dI/dV$ curves for the CFA/MgO/CoFe and Fe/MgO/Fe MTJs, respectively.

conductance curves (Fig. 5g), similar to those measured in MTJ with polycrystalline or amorphous tunnel barriers. Then, the standard free-electron model applies perfectly and the analysis using the Brinkman model [21] gives barrier parameters in agreement to those obtained from HRTEM analysis.

In contrast, one can clearly observe (Fig. 5d,f) that in case of the MgO grown on Fe, the lower period of misfit dislocation will allow a significant density of coherent tunneling channels (CH2). The tunneling spectroscopy experiments in these systems demonstrate the coherent tunneling with $k$ conservation via the complex features in the $dI/dV$ (V) curves demonstrating spin and symmetry filtering effects. The presence of local minima at RT in $G_P(T)$ validates the symmetry filtering effects by the activation of $\Delta_5$ symmetry channel at energies below 0.2eV (Fig. 5h), features which becomes even more complex at 10K [17].

This correlated structural-transport analysis in CFA/MgO and reference Fe/MgO MTJs demonstrates unambiguously the absence of coherent tunneling channels in our Heusler based junctions with thick MgO barrier. In these systems the coherence of the electronic Bloch functions in the single crystal CFA electrode is destroyed both along the propagation direction by the oblique dislocation network within the thick barrier and in the plane of the MTJ stack due to the large density of the misfit dislocations. Moreover, this reduction of the lateral (in-plane) coherence of the wave function in single crystal MTJs is particularly detrimental to symmetry dependent filtering in single crystal MTJs, where the attenuation rate within the insulator depends on the in-plane modulation of the tunneling Bloch wave function [22]. We can intuitively extrapolate our results and assume that even if longitudinal coherent channels (CH1) would exist (i.e. in CFA/MgO systems with thinner MgO barrier) a large density of dislocations within the CFA films (Fig. 3b) and the barrier (Fig. 3c) would have a direct effect on the reduction of lateral coherence of the Bloch function within the electrodes. This could have negative effects on symmetry dependent tunneling in the single crystal MTJs. Moreover, it would explain the relative few results in the literature concerning clear identification of symmetry dependent tunneling features [9, 10] even for the case of CFA/MgO MTJ systems showing significantly larger TMR ratios.

From our analysis, the impact of the MgO thickness grown on CFA on the density of misfit dislocations within the MgO and the absence of coherent tunneling channel is obvious. However, as mentioned previously, the precise control of the MgO barrier thickness during the MBE growth has been impossible. Our strategy to use RHEED oscillations calibrated for the MgO growth on Cr gave large error in thickness (20 ML instead of expected 12ML). In order to elaborate CFA/MgO/CoFe MTJs with better control of insulator thickness, we used the sputtering technique for depositing the entire stack. The MgO tunnel barrier was grown on top of CFA layer at room temperature by RF sputtering using a MgO polycrystalline target. The Ar pressure during deposition was set to 3.8 mTorr while the rf power density to 4 W/cm$^2$. The thickness of the MgO layer was controlled by assuming a constant deposition rate, for the given growth parameters, and varying the deposition time. The deposition rate was determined by X-Ray reflectometry measurements performed on a series of MgO/CFA(7.5nm)/MgO(t$_{MgO}$)/CoFe(5.5nm) samples.

Typical X-Ray reflectometry, together with the best fit to the experimental data are depicted in Fig. 6a. Figure 6b shows MgO layer thickness as a function of the deposition time. The data show a clear linear dependence and indicate that, at least for thicknesses above 1.85 nm, which was the lowest value that we were able to reason-
FIG. 6. (a) Low angle X-Ray reflectometry measurement data together with the best fit to the experiment (red curves) for the MgO/CFA(7.5nm)/MgO(tMgO)/CoFe(5.5nm) stacks for three different MgO layer thicknesses; (b) MgO layer thickness as a function of the deposition time.

The growth rate is constant. For the MTJ stack, we chose to deposit a MgO tunnel barrier with a thickness of 2.2 nm, value for which we can maintain a relative precise control. The 15 nm CoFe top magnetic electrode of the MTJ was grown on the MgO barrier by DC sputtering from a stoichiometric target. The Ar pressure during deposition was set to 1.5 mTorr and the deposition rate at 0.1 nm/s. The as-deposited CFA/MgO/CoFe stack has been annealed at 450°C to improve the crystal structure. However, as we observed from magneto-transport in patterned junctions correlated with crystallographic analysis, the annealing of the entire MTJ sputtered stack at 450°C seems to be insufficient for a complete (001) crystallization of the MTJ stack.

Magneto-transport analysis performed in these junctions shows a ten times larger TMR (Fig. 7a) than the one measured in our MBE grown samples (maximum 28% at RT). This would indicate a significant improvement of the MgO barrier quality in this case. However, the TMR value remains low with respect to the record values reported in literature in sputtered grown CFA/MgO/CoFe MTJs. This, correlated to quasi parabolic featureless dI/dV curves (insert in Fig. 7a) indicates that the coherent transport regime with symmetry and k selection, specific to single crystal MTJ, has not been achieved yet. Analysis of these results drive us to the conclusion that the crystallization of the MTJ stack at 450°C is incomplete and that the tunnel transport is yet mainly dominated by the conduction channels related to the polycrystalline zones in barrier and upper electrode. This is supported by the diffraction analysis: a polycrystalline phase subsists in the top CoFe electrode after the annealing (Fig. 7b). Further annealing studies should be performed in order to increase the crystallographic and chemical order in our sputtered MTJ stacks.

However, the figure of merit of our sputtered MTJs is that, even with crystallographic disorder, we demonstrate that by a better control of the barrier thickness, and therefore the density of dislocation, one can enhance the ratio of coherent tunneling, and consequently, by one order of magnitude the TMR.

In conclusion, in this study, we pointed out spin polarized tunneling properties specific to thick barrier CFA/MgO MTJs for which incoherent tunneling channels are dominant. Tunnel transport spectroscopy experiments at variable temperature correlated to HRTEM phase analysis demonstrate the negative effect of the misfit dislocations from the thick insulator on the longitudinal and lateral coherence of the propagating Bloch functions and consequently on the spin and symmetry dependent filtering efficiency. By reducing the density of defects within the barrier in fully sputtered MTJ devices, via a better control of barrier thickness, we enhanced the TMR ratio of the junctions by one order of magnitude.

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