Structural defects analysis versus spin polarized tunneling in Co$_2$FeAl/MgO/CoFe magnetic tunnel junctions with thick MgO barriers

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**Abstract**

We report on spin polarization reduction by incoherent tunneling in single crystal Co$_2$FeAl/MgO/Co$_{50}$Fe$_{50}$ magnetic tunnel junctions (MTJs). A large density of misfit dislocations in the Heusler based MTJs has been provided by a thick MgO barrier and its 3.8% lattice mismatch with the Co$_2$FeAl electrode. Our analysis implicates a correlated structural-transport approach. The crystallographic coherence in the real space has been investigated by High Resolution Transmission Electron Microscopy phase analysis. The electronic transport experiments in variable temperature, fitted with a theoretical extended-Glazman–Matveev model, address different levels of the tunneling mechanisms from direct to multi-center hopping. We demonstrate a double impact of dislocations, as extended defects, on the tunneling polarization. Firstly, the breaking of the crystal symmetry destroys the longitudinal and lateral coherence of the propagating Bloch functions. This affects the symmetry filtering efficiency of the $\Delta t$ states across the (001) MgO barriers and reduces the associated effective tunneling polarization. Secondly, dislocations provide localized states within the MgO gap. This determines temperature activated spin-conserving inelastic tunneling through chains of defects which are responsible for the one order of magnitude drop of the tunnel magnetoresistance from low to room temperature.

1. 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 symmetry dependent half metallicity of $\Delta t$ band in (001) bcc metals [1] and alloys [2,3], which in conjunction with an epitaxial (001) MgO barrier, providing symmetry dependent attenuation rates, 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 alloys 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 of electrodes and $\Delta t$ band symmetry filtering [8–11] provided by the symmetry dependent attenuation rate of the barrier. Moreover, this material is of special importance since it was shown to have a low Gilbert damping [12,13], essential for applications concerning 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. This regime corresponds to thick MgO barriers where the density of structural defects within the insulator is particularly important. We analyze and demonstrate the negative effect of extended defects within the barrier, e.g. dislocations, on the effective tunneling spin polarization. Our experimental strategy combines structural analysis and spin polarized...
tunneling transport in variable temperature. We demonstrate that, due to the large density of dislocations and their specific extended geometry relative to the barrier thickness, the lateral and the longitudinal coherence of the propagating wave function is lost within the barrier. The structural defects within the MTJ stack have been analyzed in terms of geometry and density using the ‘Geometrical Phase Analysis’ [14] of cross section High Resolution Transmission Electron Microscopy (HRTEM) images. The magneto-transport experiments in variable temperature are fitted within the theoretical extended Glazman–Matveev model [15]. This transport analysis allowed us to demonstrate that the spin-conserving inelastic tunneling through chains of localized states, associated with extended spatial defects within the tunneling barriers, is dominant. This defect-assisted transport mechanism is detrimental to the direct tunneling expected to provide large effective tunneling polarization in ideal, defect-free, MTJs.

2. Results and discussions

MTJs with the structure MgO(001)/Cr(20 nm)/CFA(40 nm)/MgO(4.2 nm)/CoFe(10 nm)/Au(10 nm), have been grown by combining two deposition techniques: sputtering and Molecular Beam Epitaxy (MBE). The Cr buffer layer and the bottom CFA electrode have been grown in a magnetron sputtering system, as described elsewhere [16]. Afterwards, the MgO/Cr/CFA stack has been transferred in an MBE chamber equipped with Reflection High Energy Electron Diffraction (RHEED) in situ analysis. Fig. 1a shows the RHEED pattern of the as-deposited CFA film. The shape of the pattern demonstrated that film is crystalline but rough within the RHEED coherence scale-length. Moreover, one can remark a diffusive background most probably related to the chemical disorder present in the as-deposited sample.

The CFA layer is flattened by annealing at 600 °C for 20 min (Fig. 1b). The reduction of the diffusive background is correlated with the improvement of the chemical ordering of the system from A2 towards the B2 phase [16]. In the MBE chamber by electron beam evaporation, an MgO barrier has been subsequently grown at 120 °C on top of this CFA layer. During the barrier growth, the pressure in the deposition chamber increased from 5 × 10⁻¹¹ Torr to 10⁻⁸ Torr due to the enhancement of oxygen partial pressure, as confirmed by in situ Quadrupole Mass Spectrometry analysis.

The growth kinetics has been carefully monitored by RHEED. Analysis of intensity oscillations (Fig. 1c) demonstrates a 3D growth mode of MgO on CFA. The oscillations are rapidly damped, in contrast to 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 (reference sample for MgO layer-by-layer growth, corresponding to the RHEED oscillations in Fig. 1c), shows that in this case the barrier grown on Cr is flat and has an almost perfect (001) orientation. The RHEED analysis of the barrier during the growth provides a first indication about a relatively poor structural quality of the insulator. This will be furthermore confirmed by the more detailed HRTEM analysis performed on the complete MTJ stack.

After the barrier deposition, the top CoFe magnetic electrode of the magnetic tunnel 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 layers crystallization improving also the surface flatness (Fig. 2b), as confirmed also by the X-ray theta–theta diffraction pattern (Fig. 2c). In order to magnetically operate the tunnel junction a hard–soft architecture is required. In our MTJ stack, the magnetic hardening of the top CoFe layer is obtained by direct exchange coupling with an additional 40 nm thick Co film epitaxially grown on CoFe in a hexagonal phase with the c-axis parallel to the film plane [17]. One expects that the annealing of the top CoFe electrode will also affect the barrier quality. To confirm this, an evaluation on the structural quality of the whole stack has been performed by HRTEM analysis on a cross-sectional specimen.

At a first glance, the HRTEM images (Fig. 3a) demonstrate a good crystalline quality and relative flat interfaces. 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 from HRTEM is delicate because it integrates the roughness profile over the analyzed section width. Measurements of the atomic plane 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 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. The plastic relaxation of MgO on CFA takes place in the early stages of growth, which explains the rapid oscillation damping of the in situ RHEED intensity oscillations (Fig. 1c).

Detailed information about structural defects such as dislocations and local strain fields can be extracted from HRTEM by performing the so-called ‘Geometrical Phase Analysis’ (GPA) procedure [14] (Fig. 3b–d). The GPA method consists of performing a Fourier Transform (FT) of a HRTEM image and selecting in its power spectrum reflections corresponding to lattice periodicities which show spatial deviations. Any spatial deviations in the real space correspond to phase shift in the reciprocal space and the latter can be much more easily highlighted. As an example, when performing the inverse Fourier Transform (IFT) of the HRTEM image in Fig. 3a and selecting the 002 reflection perpendicular to the interfaces, the lattice fringes corresponding to the (200) planes can be highlighted and discontinuities in these planes appear (arrows in Fig. 3b). They correspond to misfit dislocations. These misfit dislocations can be even better seen when calculating the phase shift of the (200) plane periodicities between regions of different structures i.e. of different spatial periodicities (Fig. 3c). The dislocations are perpendicular to the CFA/MgO interfaces with a periodicity of approximately 4.4 nm and correlated from one side of the barrier to the other. This becomes even clearer from Fig. 7b 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. When performing the local derivative of the phase image, the relative deformation of a given lattice compared to the

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**Fig. 3.** (a) HRTEM image of the CFA/MgO/CoFe trilayer with its FT inset. (b) (200) filtered image showing the discontinuities of the (200) planes across the interfaces (arrows). (c) Phase image evidencing the additional planes in the two electrodes and the associated misfit dislocations and (d) local deformation εyy = duyy/dy image.
other one can be measured (Fig. 3d) [16]. Fig. 3d shows the local
deformation $\varepsilon_{yy} = du_{yy}/dy$ in the direction parallel to the interface.
The mean deformation value within the interface (4%) corresponds
to the misfit between MgO and CFA. Fig. 3c and d indicates that the
deformation correlated with the misfit dislocations is located in
regions of about 1.5–2 nm width at each interface. From the
HRTEM analysis an important conclusion can be drawn. 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 perform magneto-transport experiments, our MTJ stacks
have been patterned by UV lithography and ion beam etching in
square MTJ devices with a lateral size from 10 to 50 $\mu$m. Fig. 4
illustrates typical tunnel magnetoresistance (TMR) curves for the
CFA/MgO/CoFe MTJ measured both at low (14 K) and at room
temperature (300 K). The first striking remark is the low TMR ratio
at room temperature (2.8%) measured in our systems. This is in
significant contrast to standard results obtained in similar systems,
but with lower barrier thickness, reported in the literature (larger
than 300% ratios at RT in CFA/MgO single crystal MTJs, 166% at RT
in textured CFA/MgO MTJs [8–11]). The room-temperature (RT)
TMR increases one order of magnitude, to about 23%, when the
temperature is lowered.

Following some results from the literature [18,19], one possible
explanation for the relative low TMR ratio would be the Cr
diffusion into the CFA during the 600 °C annealing stage. There-
fore, before going further in our analysis, we first checked and
refuted this hypothesis, in our samples. First, the diffusion of Cr
into the CFA should be accompanied by an increase of the coercive
field (Hc) of the CFA layer. This is not the case for our samples.
Specific studies performed in dedicated samples with and without
the annealing stage showed that the Hc of the CFA layer remains
low, around 8 Oe after the annealing state. To further demonstrate
that the Cr is not diffusing in the CFA, we performed detailed
analysis of the CFA layer by Auger spectroscopy (not shown here).
Within the limit of the Auger resolution, we did not observe any Cr
diffusion signature in or across the CFA layer. Thus, we could be
confident in further addressing another mechanism responsible
for the spin depolarizing.

Following the HRTEM analysis, one observes the presence of
extended structural defects in the epitaxial insulating barrier (i.e.
dislocations). Therefore, an immediate assumption may be that
these structural defects should affect in a negative manner the
spin polarized tunneling. This assumption could be considered
trivial. However, the exact mechanisms responsible for the depo-
larization by extended defects (such as dislocations) in the
insulator in single crystal magnetic tunnel junctions remain vague
and relatively poorly addressed in the literature. Therefore, our
goal within this paper has been to provide a deeper understanding
on the origin of the low tunneling polarization ratio, unusual for
single crystal MTJ devices. Moreover, the mechanisms identified
here could be easily extrapolated to any other epitaxial MTJ with
extended defects within the barrier. To fulfill our goal, we
performed tunnel transport experiments in variable temperature
in patterned MTJs. Our analysis has been focused on the

![Fig. 4. Magnetoresistance measurements performed on the MTJs at low (14 K) and
room temperature (300 K).](image)

![Fig. 6. TMR dependence on temperature.](image)

![Fig. 5. Temperature dependence of the tunnel conductance in the AP state. (b) Relative contribution of the N-LS chains to the total conductance in the AP state, and relative
contribution of total inelastic hopping ($N \geq 2$) to the total conductance.](image)
investigation of the transport channels contributing to the tunneling transport and it is performed within the framework of the extended Glazman–Matveev (GM) model [15] of spin-conserving hopping processes through chains of N localized states (LSs).

Within this GM model the conduction in the antiparallel (AP) magnetization configuration has the form: $G_{\text{AP}} = \sigma_0 + \sum \sigma_N \nu_N$, where $\sigma_N$ describes the hopping through $N \geq 2$ LS, $\sigma_0$ stands for the direct and 1-LS hopping and $\nu_N = N^{-2/(N+1)}$ is a characteristic exponent. Fig. 5a shows the dependence on the temperature of the AP conductance as well as a fit of the experimental points using the above equation. A first remark is that, in order to fit correctly the data, up to 6-LS chains were needed. The fit was done using only up to 2-LS chains in the low temperature range regime. Then, progressively with increasing the temperature range, higher order LS chains have been introduced when required, until the whole temperature range was fitted. Due to their different $T^{-N}$ signature, the conduction of the different N-LS chains can be extracted from the fit. In Fig. 5b we have plotted the weighted contribution $W_N = G_N/G_{\text{AP}}$ of each conduction channel to the total conductance in the AP state. This analysis provides important information about the tunneling contribution of different channels as a function of temperature. While, in the low temperature regime the conductance is mainly due to the 0 and 1-LS contribution (which cannot be distinguished within the extended GM model).
approach [15], when the temperature is increased towards the room temperature (RT) higher order conductivity chains (N≥2) become dominant. Within the model of spin-conserving hopping [15] the TMR assigned to a variety containing N-LS is given by TMR(N) = ((1 + P) N - (1 - P) N) / 2((1 - P) N) - 1, where βN = 1/(N + 1) and P is the effective temperature dependent spin polarization of electrodes P = P0(1 - aT 3/2) [20], α is the spin-wave parameter related to the interfacial Curie temperature and P0 is the effective spin polarization at 0 K. A direct result of the above relation is that even if the channels are spin conserving, the TMR gradually decreases when N is increased. The total TMR is given by the sum of the TMR of each conduction variety weighted by their fractional contribution TMR = \sum N W(N) × TMR(N).

Fig. 6 shows the evolution of the TMR with temperature as well as a fit using the above described model. As can be seen from the figure, the fit reproduces well the experimentally observed strong TMR decrease with the temperature. In our fitting procedure, due to the large thickness of the barrier and confronting with phase images extracted from HRTEM (see later Fig. 7b and e and related discussion), we reasonably ignored the direct channel and took into account only the resonant tunneling with one (N = 1) and higher order hopping channels (N≥2). Consequently, our extended GM analysis provided an effective polarization of P0 = 0.57. This is in agreement with the theoretical and experimental expected values for CFA films showing B2 disorder [21–23]. Therefore, one can conclude that despite standard electrode polarization provided by our CFA films with B2 disorder, the low temperature TMR is reduced from 0.96, as expected for direct tunneling (N = 0), to 0.23 due to the dominant resonant tunneling (N = 1) via localized levels associated with extended structural defects within the thick insulator. Furthermore, the fact that the value of the effective spin polarization of P0 = 0.57 is in agreement with the theoretical and experimental expected values for CFA films showing B2 disorder indicates that our thick barrier MTJs do not benefit from the Δ1 band symmetry filtering of the MgO barrier, which should increase the effective tunneling spin polarization to higher values [8–11]. This is a direct consequence of the dominating incoherent hopping assisted transport in our thick barrier CFA/MgO/CoFe MTJs. To further confirm this we performed detailed structural investigations by HRTEM phase image contrast focused on the MgO barrier zone, correlated with tunneling spectroscopy experiments (dI/dV curves) measured in patterned MTJ devices. The results (Fig. 7a,b,g) were compared to those obtained in standard Fe/MgO/Fe MTJs (Fig. 7c,d,h) where we have previously demonstrated that tunneling coherent spin and symmetry dependent tunneling channels dominate the conductance and are responsible for the large tunneling polarization and TMR [17,24,25]. Fig. 7b illustrates the phase shift image corresponding to the real space image of Fig. 7a showing the presence of the misfit dislocations inside the thick MgO barrier grown on (001) CFA. The density of dislocations is significantly larger than the one corresponding to the MgO barrier grown on (001) Fe (Fig. 7d). Consequently, as sketched in Fig. 7e and f, 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 channels are present. Therefore, the tunneling transport in the case of the thick MgO barrier will be dominated by incoherent hopping tunneling channels (denoted CH2) associated with dislocations and lattice deformations (Figs. 7b and d) within the thick MgO barrier. These incoherent hopping transport channels are temperature activated and, as demonstrated from extended GM analysis, they can explain the strong temperature dependence of the conductance and TMR in our thick MgO barrier CFA based MTJs. The signature of the incoherent inelastic transport channels is also suggested by the parabolic dynamic conductance curves (Fig. 7g), similar to those measured in MTJs with polycrystalline or amorphous tunnel barriers. This also demonstrates that even in MTJs with crystalline electrodes, when the structural coherence is lost across the stack and coherent direct tunneling is quenched detrimental to incoherent hopping assisted transport, the fine signature of electrode density of states becomes smeared within the parabolic dominant shape of the dI/dV curves. The standard free-electron model applies fairly (not shown here) and the fit using the Brinkman model [26] gives barrier parameters in agreement with those obtained from HRTEM analysis.

The analysis performed on CFA/MgO/CoFe MTJs has been confronted to similar analysis performed on standard Fe/MgO/Fe MTJs known to provide efficient spin and symmetry filtering [17,24,25]. In contrast to CFA/MgO/CoFe MTJs, one can clearly observe (Fig. 7d and f) that in the case of the MgO grown on Fe, the lower corresponding period of misfit dislocation will allow a significant density of direct coherent tunneling channels (CH2). The tunneling spectroscopy experiments in these control systems demonstrate the coherent tunneling with k conservation (enabled by structural coherence across the stack) via the complex features in the dI/dV(V) curves demonstrating spin and symmetry filtering effects. As we previously argued [24], the presence of local minima event at RT in Gk(T) measured in Fe/MgO/Fe MTJs validates the symmetry filtering effects by the activation of additional Δs symmetry channel (Fig. 7h) [17,24]. Similar temperature dependent transport analysis involving the extended Glazman–Matveev model has been performed for Fe/MgO/Fe MTJ (Fig. 8). The purpose of this analysis was to give further proof of our hypothesis concerning the dominant direct coherent tunneling with respect to the spin-conserving hopping processes in Fe/MgO/Fe MTJ, compared with the case of CFA/MgO/CoFe MTJs, where incoherent transport channels are dominant (Fig. 7e).

Fig. 8a shows the dependence of the AP conductance and the corresponding fit within the GM model applied to Gα(N,T) curves.

**Fig. 8.** (a) Temperature dependence of the tunnel conductance in the AP state. (b) Relative contribution of the N-LS chains to the total conductance in the AP state.
measured in Fe/MgO/Fe MTJs. In contrast to the CFA/MgO/CoFe MTJs, here the experimental data are correctly fitted only implying accumulation up to 3-LS chains. Fig. 8b illustrates the relative contribution of each conduction channel to the total conductance in the AP state. Although 2 inelastic hopping conduction channels are thermally activated the 0-LS and 1-LS remain dominant even at room temperature, as expected from the HRTEM analysis and tunneling spectroscopy experiments. The fit of the TMR dependence on temperature (not shown here) gives an effective polarization of $P_{\text{eff}} = 0.84$, as expected for a (001) Fe electrode in conjunction with a (001) MgO barrier [2], which benefits from $\Delta_1$ band symmetry filtering properties provided by the symmetry dependent attenuation rate of the barrier. Moreover, our analysis performed on Fe/MgO/Fe MTJs emphasizes the negative role of dislocations in these systems on the tunneling spin polarization amplitude. Even if larger TMR ratios are measured at RT in these systems, the value of the experimental TMR is lower than the theoretical predictions. The reduction of TMR has been previously determined. This is an agreement with the large 180% TMR in the case of reference Fe/MgO systems, where the coherent tunneling polarization. Related to the quenching of the crystal symmetry coherence across the MTJ stack, 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 misfit dislocations. Moreover, this reduction of the lateral 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 [27]. Our analysis provides a reasonable effective spin polarization of 0.57 at OK for our CFA/MgO-thick system which confirms the absence of $\Delta_1$ band symmetry filtering within the thick MgO barrier. Beyond the direct effect on symmetry filtering with consequences on effective tunneling polarization amplitude, we demonstrate here that the dislocations may have a much larger negative impact on the tunneling magnetoresistance amplitude. In our MTJs with thick MgO barrier and large density of dislocations the transport in variable temperature experiments combined with HRTEM phase analysis demonstrates the dominance of thermally activated inelastic hopping channels. This explains the relative low values of the TMR and its strong decrease in temperature. In contrast, in the case of reference Fe/MgO systems, where the coherent channels dominate, an effective spin polarization of 0.84 has been determined. This is an agreement with the large 180% TMR measured in these systems at RT.

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