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Room temperature spin filtering in epitaxial cobalt-ferrite tunnel barriers

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We report direct experimental evidence of room temperature spin filtering in magnetic tunnel junctions (MTJs) containing CoFe_2O_4 tunnel barriers via tunneling magnetoresistance (TMR) measurements. $\text{Pt}(111)/\text{CoFe}_2\text{O}_4(111)/\gamma\text{-Al}_2\text{O}_3(111)/\text{Co}(0001)$ fully epitaxial MTJs were grown in order to obtain a high quality system, capable of functioning at room temperature. Spin polarized transport measurements reveal significant TMR values of -18% at 2 K and -3% at 290 K. In addition, the TMR ratio follows a unique bias voltage dependence that has been theoretically predicted to be the signature of spin filtering in MTJs containing magnetic barriers. CoFe_2O_4 tunnel barriers therefore provide a model system to investigate spin filtering in a wide range of temperatures.

The generation of highly spin-polarized electron currents is one of the dominant focusses in the field of spintronics. For this purpose, spin filtering is one very interesting phenomenon, both from a fundamental and from a technological stand point, that involves the spin-selective transport of electrons across a magnetic tunnel barrier. Successful spin filtering at room temperature could potentially impact future generations of spin-based device technologies [1, 2] not only because spin filters may function with 100% efficiency [3], but they can be combined with any non-magnetic metallic electrode, thus providing a versatile alternative to half-metals or MgO-based classic tunnel junctions.

The spin filter effect originates from the exchange splitting of the energy levels in the conduction band of a magnetic insulator. As a consequence, the tunnel barrier heights for spin-up and spin-down electrons ($\Phi_{\uparrow(\downarrow)}$) are not the same, leading to a higher probability of tunneling for one of the two spin orientations : $J_{\uparrow(\downarrow)} \propto \exp(-\Phi_{\uparrow(\downarrow)}^{1/2}t)$, where t is the barrier thickness. The spin filter effect was first demonstrated in EuS using a superconducting electrode as a spin analyzer (i.e. Merservey-Tedrow technique) [4], and has since been observed in EuSe [3] and EuO [5] by this method. Because the Merservey-Tedrow technique is limited to low temperatures, tunneling magnetoresistance (TMR) measurements in magnetic tunnel junctions (MTJs) [6] have been used to show the spin filter capability of higher temperature spin filters such as BiMnO₃ [7] and NiFe₂O₄ [8]. However, no TMR effects are currently reported from any spin filter at room temperature.

CoFe₂O₄ is a very promising candidate for room temperature spin filter applications thanks to its high Curie temperature ($T_C = 793$ K) and good insulating properties. Electronic band structure calculations from first principles methods predict CoFe₂O₄ to have a band gap of 0.8 eV, and an exchange splitting of 1.28 eV between the minority (low energy) and majority (high energy) levels in the conduction band [9] (see Fig.3-b), thus confirming its potential to be a very efficient spin filter, even at room temperature. Recently, a tunneling spectroscopy study of CoFe₂O₄/MgAl₂O₄/Fe₃O₄ double barrier tunnel junctions revealed optimistic results for the spin-filter efficiency of CoFe₂O₄ [10]. However, the polarization (P) and TMR values obtained in this work were indirectly extracted from a complex model developed to fit experimental current-voltage curves rather than from direct Merservey-Tedrow or TMR measurements.

In order to accurately demonstrate the spin filtering capabilities of CoFe₂O₄ up to room temperature, we have prepared CoFe₂O₄(111)/ γ -Al₂O₃(111)/Co(0001) fully epitaxial tunnel junctions by oxygen plasma-assisted molecular beam epitaxy (MBE) on Pt(111) underlayers. The details of the sample growth process are published elsewhere [11]. In this system, the spinel γ -Al₂O₃ serves to decouple the CoFe₂O₄ and Co magnetic layers. Before any spin-polarized transport measurements were performed on the full MTJ system, our CoFe₂O₄(111)/ γ -Al₂O₃(111) tunnel barriers were carefully characterized by a wide range of techniques in order to optimize their structural and chemical properties [11]. Fig. 1 shows a high resolution transmission electron microscopy (HRTEM) study demonstrating the high crystalline quality of our CoFe₂O₄ (5 nm)/ γ -Al₂O₃ (1.5 nm)/Co (10 nm) multilayers. In particular, we observe near perfect epitaxy in the single crystalline CoFe₂O₄(111)/ γ -Al₂O₃(111) tunnel barrier, which is a consequence of the optimized growth conditions and the spinel structure of both constituents.

The magnetic properties of a CoFe₂O₄/ γ -Al₂O₃ tunnel barrier were measured at room temperature and at 4 K,

yielding coercivities (H_c) of 220 Oe and 500 Oe respectively (not shown). We also observed a rather weak remanent magnetization around 40% and lack of saturation and magnetic reversibility for fields as high as 5 T at 4 K. These properties are common for spinel ferrite thin films and have been attributed to the presence of antiphase boundaries [12]. We note the magnetic properties of the full Pt/CoFe₂O₄/γ-Al₂O₃/Co MTJs were also characterized in order to verify the magnetic decoupling of the CoFe₂O₄ and Co layers, necessary for TMR measurements [11].

Spin filter tunnel junctions were patterned by advanced optical lithography. Spin-polarized transport measurements were carried out in the two probe configuration, as the room temperature junction resistance was 3 orders of magnitude higher than that of the Pt cross strip and contacts. Fig.2-a,b clearly demonstrates the TMR effect in a Pt/CoFe₂O₄/γ-Al₂O₃/Co tunnel junction both at 2 K and 290 K. This result is direct experimental evidence of the spin filter effect in CoFe₂O₄ at low temperature and *at room temperature*. At 2 K we calculate a TMR value of -18% using the relation $TMR = (R_{AP} - R_P)/R_P$ where R_{AP} and R_P are the resistance values in the antiparallel and parallel magnetic configurations. At room temperature, $TMR = -3\%$. The abrupt drop in the TMR curve at ±200 Oe corresponds to the switching of the Co electrode, while the gradual increase back to ±6 T agrees with the progressive switching and lack of saturation in CoFe₂O₄ seen in the magnetization measurements. The negative sign of the TMR indicates that the CoFe₂O₄ spin filter and Co electrode are oppositely polarized which is most consistent with the negative P predicted for CoFe₂O₄ in band structure calculations [9], and the positive P measured for Co by the Meservey-Tedrow technique. Taking $P_{Co}=40\%$ [13], we may approximate $P_{CoFe_2O_4}$ from Jullière's formula: $TMR = 2P_1P_2/(1 - P_1P_2)$ where $P_1=P_{Co}$ and $P_2=P_{CoFe_2O_4}$ [14]. This gives $P_{CoFe_2O_4}=-25\%$ at 2K and $P_{CoFe_2O_4}=-4\%$ at room temperature. Due to the low remanence in our CoFe₂O₄ films, one could expect these values to increase significantly with the future improvement of their magnetic properties, reaching 50% or higher. The considerable decrease of $P_{CoFe_2O_4}$ at high temperature could be explained by the thermal excitation of the spin up electrons into their corresponding majority spin conduction band, if the conduction band splitting were small with respect to $k_B T$.

The $I - V$ characteristics representative of the Pt/CoFe₂O₄/γ-Al₂O₃/Co system are shown in Fig.2-c. Analyzing the second derivative of these tunneling spectroscopy measurements, we obtain a good estimation of Φ from the bias voltage at which these deviate from linearity. Fig.2-d clearly shows that d^2I/dV^2 is linear for $-60 \text{ mV} < V < 60 \text{ mV}$, indicating direct tunneling in this regime. We also obtain the same value of Φ from the $(dI/dV)/(I/V)$ characteristics which show a peak at 60 mV corresponding to the onset of the conduction band. We note that this Φ value does not account for the voltage drop across the γ-Al₂O₃ barrier, which should in fact lower it further. In either case, the relatively small tunnel barrier height is quite consistent with the electronic band structure calculations schematized in Fig.3-b, which predict CoFe₂O₄ to have a small electronic band gap and an intrinsic Fermi level that is close to the first level of the conduction band.

In addition, the $I - V$ curves taken in the antiparallel (±0.08 T) and parallel (∓6 T) states may be used to extract the TMR bias dependence by using the definition $TMR = (I_P - I_{AP})/I_{AP}$. The result, shown in Fig.3-a, is a steady increase in absolute value of the TMR with increasing V up to a certain value, followed by a slight decrease for higher biases. This exact behavior was theoretically predicted by A. Saffarzadeh to be the signature of spin filtering in MTJs

containing a magnetic barrier [15], and has only recently been observed experimentally with EuS at low temperature [16]. However, it has never been verified in higher T_C magnetic oxide tunnel barriers [7, 8]. The fact that the TMR increases with increasing V both at low temperature and at room temperature proves that the TMR we observe truly results from spin filtering across the CoFe_2O_4 barrier. The onset of spin filtering, given by the region for which TMR increases with V , may be identified from the low temperature curve in Fig.3-a around ± 30 mV and persists until $+130$ mV (-100 mV) for positive (negative) bias.

Quantitative comparison of the spin filter regime in our $\text{TMR}(V)$ curves with the calculations of Szotek *et al.* yields an exchange splitting (in the tens of meV in our junctions) that is significantly lower than that predicted for the inverse spinel structure (1.28 eV). This observation is consistent with the temperature sensitivity of the TMR measurement discussed earlier. The electronic band structure of the CoFe_2O_4 barrier is likely influenced by the presence of structural and/or chemical defects, many of which are difficult to account for in model systems for such calculations. The presence of Co^{3+} , for example, is one defect that has been predicted by Szotek *et al.* [9] to reduce the band gap as well as the conduction band splitting while favoring an energetically stable state. Furthermore, one can not ignore the possible influence of oxygen vacancies or antiphase boundaries, as these are known to influence the magnetic order in spinel ferrites [12], and thus likely the exchange splitting as well. Further studies are underway to better quantify the effects of both structural and chemical defects on the spin polarized tunneling across CoFe_2O_4 .

In summary, we have demonstrated room temperature spin filtering in fully epitaxial $\text{Pt}(111)/\text{CoFe}_2\text{O}_4(111)/\gamma\text{-Al}_2\text{O}_3(111)/\text{Co}(0001)$ MTJs where CoFe_2O_4 was the magnetic tunnel barrier. TMR values of -18% and -3% were observed at 2 K and 290 K respectively. Furthermore, the experimental TMR ratio increased with increasing bias voltage, reproducing the theoretically predicted behavior for a model spin filter system. The similarity between our experimental $\text{TMR}(V)$ curves and those previously predicted in the literature not only proves the spin filtering capability of CoFe_2O_4 , but also validates the theoretical and phenomenological models describing spin-polarized tunneling across a magnetic insulator.

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FIGURE CAPTIONS

FIG. 1: HRTEM image of a CoFe_2O_4 (5 nm)/ $\gamma\text{-Al}_2\text{O}_3$ (1.5 nm)/Co (10 nm) trilayer deposited directly on a sapphire substrate, and showing the exceptional quality of the fully epitaxial system.

FIG. 2: TMR as a function of applied magnetic field for a Pt(20 nm)/ CoFe_2O_4 (3 nm)/ $\gamma\text{-Al}_2\text{O}_3$ (1.5 nm)/Co(10 nm) tunnel junction at 2 K (a) and at room temperature (b) with an applied bias voltage of 200 mV. The junction area A was $24 \mu\text{m}^2$. A zoom of the Co switching at room temperature is shown in the insert of (b). The $I - V$ characteristics, and d^2I/dV^2 fitted linearly for $-\Phi \leq V \leq \Phi$ are shown in (c) and (d).

FIG. 3: (a) TMR as a function of bias voltage for a Pt(20 nm)/ CoFe_2O_4 (3 nm)/ $\gamma\text{-Al}_2\text{O}_3$ (1.5 nm)/Co(10 nm) tunnel junction at 2 K and 300 K. The open data points correspond to TMR values obtained from $R(H)$ measurements. (b) Schematic representation of the CoFe_2O_4 band structure based on first principles studies [9].

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