Large Inverse TMR in $Co_2Cr_{0.6}Fe_{0.4}Al$ Based Magnetic Tunnel Junctions

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Magnetic tunnel junctions based on the Heusler alloy $Co_2Cr_{0.6}Fe_{0.4}Al$ and MgO barriers fabricated by magnetron sputtering exhibit a large inverse tunneling magnetoresistance (TMR) effect of up to -66% at room temperature. The largest value of -84% at 20 K reflects a rather weak influence of temperature. The dependence on the voltage drop shows an unusual behavior with two almost symmetric peaks at ± 600 mV with large inverse TMR ratios and small positive values around zero bias. These results are of high relevance for applications, as they combine a large TMR ratio at a high output voltage with a moderate temperature dependence.

The tunnel magnetoresistance effect (TMR) is subject of intense research due to its potential for spintronic applications. The aim is to achieve a large TMR ratio at practical output voltages in combination with a weak temperature dependence. A promising strategy for obtaining large TMR effects is to use ferromagnetic (FM) electrodes with an *intrinsically* high spin polarization, such as half-metallic ferromagnets featuring 100% spin polarization of the carriers. Among the vast family of the half-metallic ferromagnets, the Heusler alloys are promising candidates due to their high Curie temperatures well above 300 K. In the last years, experimental efforts were concentrated on improving the structure of Heusler thin films and the properties of the interface between the Heusler electrode and the oxide barrier. Relatively high TMR ratios up to 90% at room temperature (RT) have been obtained using Co-based full-Heusler alloy thin films, in particular $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA) [1]. Here we report on large *inverse* TMR values obtained from CCFA/MgO/$Co_{80}Fe_{20}$ magnetic tunnel junctions (MTJ) [2].

Details on the preparation and characterization of $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA) Heusler thin films have been published elsewhere [2, 3]. In contrast to previous reports about the growth of CCFA films on MgO(001) substrates[1, 4], our CCFA films are grown at RT and adopt a different crystalline orientation with respect to the MgO(001) substrate [3]. The [011] direction of the CCFA films is parallel to [001] of the MgO(001) substrate. The CCFA films have the B2 structure, since the (111) reflection is absent in the X-ray diffraction patterns. Magnetization measurements of extended CCFA films reveal ferromagnetic ordering with Curie temperatures up to 630 K after annealing in vacuum at 773 K. The total magnetic moment found is about $2.5\mu_B$ per formula unit (f.u.). This value is small compared to the theoretical bulk value of $3.8\mu_B$/f.u., but still comparable to the moments reported in Ref. [4] for films grown at elevated temperature.

MTJs are prepared by magnetron sputtering at RT without breaking the vacuum with the layer sequence MgO(100) / MgO(40 nm) / CCFA(25 nm) / MgO(3 nm) / $Co_{80}Fe_{20}$(5 nm) / IrMn(15 nm). A 40 nm-thick MgO seed layer is deposited on the MgO substrate to improve the texture of the CCFA electrode. CCFA and MgO are deposited by DC and RF stimulated discharge, respectively, from stoichiometric targets. The completed stack is annealed *in-situ* for 1 hour at 523 K in order to improve the interface quality.

Junctions with an area from 3×3 up to 15×15 μm^2 with cross-bar electrodes are patterned for magnetotransport measurements in the current-perpendicular-plane (CPP) geometry by optical lithography. The transport measurements are performed with a DC setup in the standard 4-point geometry using a constant current source. $I-V$ characteristics measured at RT in zero field show nonlinear, *i.e.* non-Ohmic behavior.

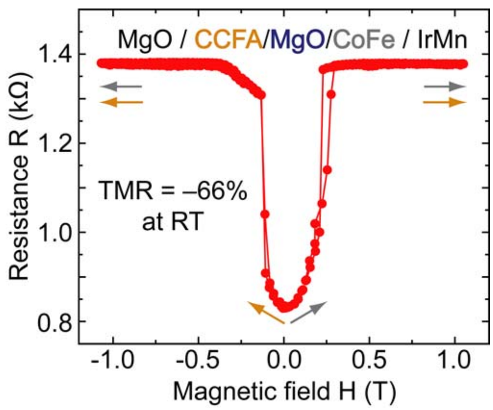

FIG. 1: TMR curve of a CCFA(25 nm)/MgO(3 nm)/ $Co_{80}Fe_{20}$(5 nm) MTJ measured at RT.

In Fig. 1 we present a magnetoresistance curve measured at RT on a CCFA/MgO/$Co_{80}Fe_{20}$ MTJ deposited on MgO(100) substrates with a 40 nm-thick MgO seed layer. Obviously, we observe an *inverse* TMR effect. The TMR ratio defined as

TMR = $(R_{AP} - R_P)/R_{AP}$, where R_{AP} is the smallest resistance value in the antiparallel magnetization configuration and R_P denotes the highest resistance in the saturated state, reaches -66%. This TMR value at RT is relatively large for structures comprising a FM Heusler electrode. The bell-shaped MR curve depicted in Fig. 1 suggests a noncollinear orientation of the layers magnetizations for $H = 0$ due to magnetic coupling of the two FM layers (arrows in Fig. 1). This is also confirmed by SQUID measurements, where independent switching of the two electrodes is found to be hindered. Antiferromagnetic, Néel-type and biquadratic coupling due to interface roughness are the most likely coupling mechanisms.

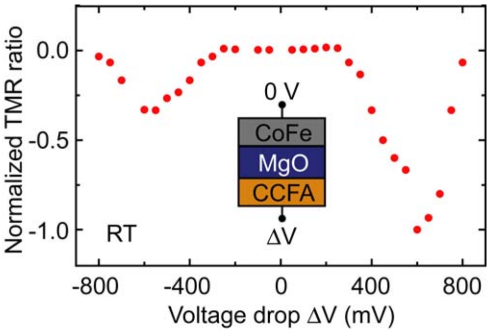

FIG. 2: Normalized TMR ratio of a CCFA(25 nm)/MgO(3 nm)/$Co_{80}Fe_{20}$(5 nm) MTJ as a function of the voltage drop ΔV across the junction measured at RT.

Figure 2 shows the typical dependence of the TMR ratio on the voltage drop ΔV across the MTJ measured in the parallel configuration at $H = \pm 1$ T. ΔV is experimentally controlled by varying the current bias supplied by the constant current source and is defined with respect to the $Co_{80}Fe_{20}$ electrode (see inset of Fig. 2).

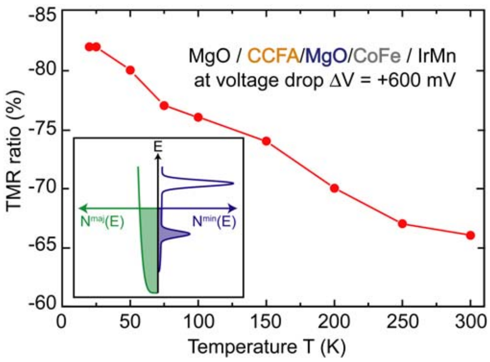

FIG. 3: Temperature dependence of the TMR ratio for a CCFA(25 nm)/MgO(3 nm)/$Co_{80}Fe_{20}$(5 nm) MTJ at $\Delta V = +600$ mV. Inset: Schematic spin-split DOS of CCFA.

In Fig. 3 we plot the temperature dependence of the TMR ratio measured at $\Delta V = +600$ mV. In contrast to MTJs with other Heusler electrodes, our MTJs show only a moderate temperature dependence. The TMR ratio increases from -66% at RT to -84% upon cooling down to 20 K. Similar behavior was reported in Ref. [5] for fully epitaxial CCFA/MgO/CoFe MTJs.

The most striking features of our results are (i) the dominant large *inverse* TMR, (ii) its strong dependence on the voltage drop ΔV, and (iii) the weak temperature dependence. The strong variation of the TMR ratio with the voltage drop, which clearly deviates from the usually found cusp-like behavior, and the weak temperature dependence suggest a strong influence of the DOS on the TMR. In the framework of Jullière's model, the TMR ratio can only be negative when the effective spin polarizations P_L and P_R on the left and right side of the barrier, respectively, are of opposite sign. Sharp features in the spin-split DOS of the electrodes give rise to a bias dependence of the effective polarizations and thus the TMR ratio. The nature of the bonding at the ferromagnet-insulator interface can influence the character of the tunnelling electrons and thus both size and sign of the effective polarization. Due to the preferential (110) orientation of our CCFA films, the bonding at the CCFA/MgO interface in our TMR structures is significantly different from the commonly found (100) orientation. The related differences in the band structures could be the reason for the inverse TMR ratios for certain ΔV in our experiment. Assuming that CCFA shows much sharper features in the spin-split DOS than $Co_{80}Fe_{20}$, *e.g.* due to a (pseudo-) gap and band edges, the data in Fig. 2 can be qualitatively explained by the schematic spin-split DOS of CCFA in the inset of Fig. 3. The large interval of 300-400 mV between the Fermi level and the onsets of the peaks in the model DOS of CCFA explains the much weaker temperature dependence than found for other systems.

In conclusion, we observed a large inverse TMR effect in magnetron sputtered CCFA/MgO/$Co_{80}Fe_{20}$ MTJs at RT. The TMR ratio shows an unusual dependence on the voltage drop ΔV across the structure with large negative values of up to -66% at $\Delta V = \pm 600$ mV and small positive values around zero bias. The temperature dependence is moderate with an increase from -66% to -84% ($\Delta V = +600$ mV) upon cooling from RT to 20 K. We proposed that these findings are related to density-of-states effects of the CCFA electrode or the CCFA/MgO interface. From the application point of view, our results are of high relevance, as they combine a large TMR ratio at a relatively high output voltage with a moderate temperature dependence.

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