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Large inverse tunneling magnetoresistance in $Co_2Cr_{0.6}Fe_{0.4}Al/MgO/Co_{80}Fe_{20}$ magnetic tunnel junctions

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Magnetic tunnel junctions with the layer sequence $Co_2Cr_{0.6}Fe_{0.4}Al/MgO/Co_{80}Fe_{20}$ were fabricated by magnetron sputtering at room temperature (RT). The samples exhibit a large inverse tunneling magnetoresistance (TMR) effect of up to −66% at RT. 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. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2728034]

The tunnel magnetoresistance (TMR) effect, i.e., the resistance change upon application of a magnetic field in a structure consisting of two ferromagnetic (FM) layers separated by a thin insulating barrier, is a subject of intense experimental research due to its potential for spintronic applications.[1] The aim is to achieve a large TMR ratio for practical output voltages in combination with a weak temperature dependence. Recently, very high TMR ratios exceeding 200% at room temperature (RT) have been reported for magnetic tunnel junctions (MTJs) with fully epitaxial or highly oriented MgO barriers and Fe,[2] CoFe,[3] and CoFeB (Ref. 4) electrodes. Resonant tunneling and specific features in the band structures of MgO- and Fe-based alloys with bcc structure give rise to a spin-filter effect, such that predominantly highly spin-polarized states of the FM contribute to the tunneling current.[5] Weak contributions from other states and any breaking of the translational invariance parallel to the interfaces (e.g., interface roughness, defects) limit the maximum achievable TMR ratio.[6] Another strategy for obtaining large TMR effects is to use FM electrodes with an *intrinsically* high spin polarization, such as half-metallic ferromagnets featuring 100% spin polarization of the carriers.[7] 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.[8]

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 have been obtained using Co-based full-Heusler alloy thin films, e.g., Co_2MnSi (Refs. 9 and 10) and $Co_2Cr_{0.6}Fe_{0.4}Al$.[11–13] Marukame *et al.*[14] prepared fully epitaxial $Co_2Cr_{0.6}Fe_{0.4}Al/MgO/CoFe$ MTJs with up to 90% TMR at RT. In this letter we report on large *inverse* TMR values obtained from $Co_2Cr_{0.6}Fe_{0.4}Al/MgO/Co_{80}Fe_{20}$ MTJ structures.

Details on the preparation and characterization of $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA) Heusler thin films have been published elsewhere.[15] In contrast to previous reports about the growth of CCFA films on MgO(001) substrates,[13,16,17] our CCFA films are grown at RT and adopt a different crystalline orientation with respect to the MgO(001) substrate.[15] The [011] direction of the CCFA films is parallel to [001] of the MgO(001) substrate. The CCFA films have the *B*2 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 $2.5\mu_B$/f.u. This value is small compared to the theoretical bulk value of $3.8\mu_B$/f.u.,[18] but still comparable to the moments reported in Refs. 16 and 17 for films grown at elevated temperature.

MTJs are prepared by magnetron sputtering at RT without breaking the vacuum with the following 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 discharges, respectively, from stoichiometric targets. The sputtering rates monitored with a quartz balance are calibrated by measuring the thicknesses of individual thin film layers by x-ray reflectivity. The completed stack is annealed *in situ* for 1 h at 523 K in order to improve the interface quality. However, we cannot apply a magnetic field during the *in situ* annealing. Thus, the $Co_{80}Fe_{20}$ electrode is not exchange biased. A structural characterization of the 3-nm-thick MgO layer could not be performed. Hence, we have no information about a possible texture of the MgO barrier. 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 geometry by optical lithography. The transport measurements are performed with a dc setup in the standard four-point geometry using a constant current source. *I-V* characteristics measured at RT in zero field show a nonlinear, i.e., non-Ohmic, behavior.

In Fig. 1 we present a magnetoresistance curve measured at RT on a CCFA/MgO/$Co_{80}Fe_{20}$ MTJ deposited on MgO(100) substrate with a 40-nm-thick MgO seed layer. Obviously, we observe an inverse TMR effect. The TMR ratio defined as in Ref. 19 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%. The minimal resistance times area product is about 80 kΩ μm^2. The junction resistances are 1 kΩ or larger in all samples and,

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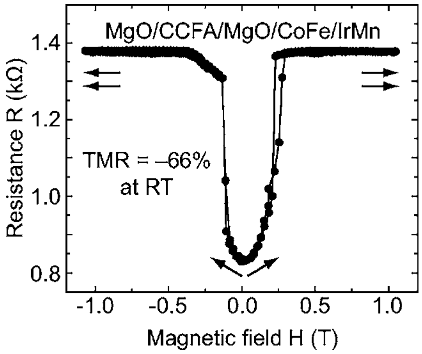

FIG. 1. Magnetoresistance curve of a 10×10 μm^2 MTJ with layer sequence MgO/MgO (40 nm)/CCFA (25 nm)/MgO (3 nm)/$Co_{80}Fe_{20}$ (5 nm)/IrMn (15 nm). The measurement is performed at RT and yields an inverse magnetoresistance of −66% for $\Delta V = +600$ mV.

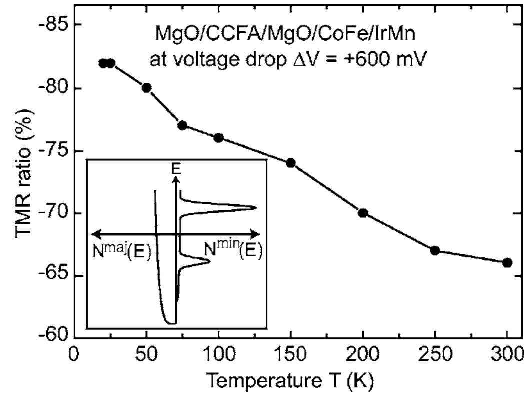

FIG. 3. Temperature dependence of the TMR ratio for a MgO/MgO (40 nm)/CCFA (25 nm)/MgO (3 nm)/$Co_{80}Fe_{20}$ (5 nm)/IrMn (15 nm) MTJ measured at a voltage drop $\Delta V = +600$ mV. Inset: schematic spin-split DOS of CCFA.

thus, clearly exceed the lead and contact resistances of 10–20 Ω. Therefore, there is no geometrical enhancement in the TMR ratios. The TMR value obtained 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 layer magnetizations for $H=0$ due to magnetic coupling of the two FM layers (arrows in Fig. 1). This is also confirmed by superconducting quantum interference device 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.

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). The inverse TMR effect is dominant over a wide range of positive and negative ΔV. Only for $|\Delta V| < 300$ mV do we obtain a small positive TMR ratio of less than 1%. From $\Delta V = +300$ to $+600$ mV the TMR ratio increases and abruptly decreases for larger ΔV. A similar almost symmetric behavior is observed for negative ΔV, except that the peak at −600 mV is less pronounced.

In Fig. 3 we plot the temperature dependence of the TMR ratio measured at $\Delta V = +600$ mV. In contrast to MTJs with Co_2MnSi and Co_2MnGe Heusler electrodes,[9,20,21] 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. 21 for fully epitaxial CCFA/MgO/CoFe MTJs.

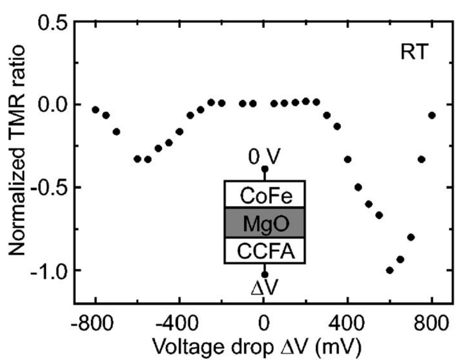

FIG. 2. Normalized TMR ratio of a MTJ with layer sequence MgO/MgO (40 nm)/CCFA (25 nm)/MgO (3 nm)/$Co_{80}Fe_{20}$ (5 nm)/IrMn (15 nm) as a function of the voltage drop ΔV across the junction measured at RT. Inset: ΔV is defined with respect to the $Co_{80}Fe_{20}$ electrode.

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. They have been observed in many MTJs prepared in different runs (e.g., Figs. 1 and 2 show data from different runs). The inverse sign and the dependence on voltage drop are similar in all cases, only the magnitude of the effect differs from junction to junction. Thomas *et al.*[22] reported on inverted spin polarization of Co_2MnSi and Co_2FeSi Heusler alloys, which gave rise to a small inverse TMR ratio of about −3% at bias voltages exceeding −1.3 V. For small bias, however, normal TMR of up to 100% was prevailing. Yamamoto *et al.*[21] also observed a crossover to small negative TMR at negative bias voltage in MTJ structures comprising a Co_2MnGe electrode. These authors concluded from the conductance measurements that the inverse TMR was due to direct tunneling and thus reflected the bulk spin-dependent electronic density-of-states (DOS) of the Co_2MnGe electrode. In our case, the strong variation of the TMR ratio with the voltage drop, which clearly deviates from the usually found cusplike behavior, and the weak temperature dependence also 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 sides 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.[23] It was shown (see Refs. 19, 24, and 25) that the nature of the bonding at the ferromagnet-insulator interface can influence the character of the tunneling 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 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. We have also assumed that the effective polarization of the $Co_{80}Fe_{20}$/MgO interface is positive. If it is negative, minority and majority spin directions in the inset of Fig. 3 must be exchanged. 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 those found for other systems.[20] Theoretical studies of the spin-split DOS for CCFA/MgO(110) interfaces and spin-dependant transport as finite voltages beyond the Jullière model could give more insight into the origin of the large inverse TMR effect. In particular, such calculations could also properly take into account the spin and bias dependence of the transmission probabilities, which have been shown to strongly affect the TMR effect.[26]

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 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. Further investigations, both experimental and theoretical, are needed in order to understand and control the large inverse TMR as well as its bias and temperature characteristics. 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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