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Large tunneling magnetoresistance effect at high voltage drop for Co-based Heusler alloy/MgO/CoFe junctions

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Growth and magnetic characterization of thin films of $Co_2Cr_{0.6}Fe_{0.4}Al$ and Co_2MnSi full-Heusler compounds are investigated. Thin films were deposited by magnetron sputtering at room temperature directly onto oxidized Si wafers. These Heusler films are magnetically very soft and ferromagnetic with Curie temperatures well above room temperature. Polycrystalline $Co_2Cr_{0.6}Fe_{0.4}Al$ Heusler films combined with MgO barriers and CoFe counter electrodes are structured to magnetic tunnel junctions and yield almost 50% magnetoresistance at room temperature. The magnetoresistance shows a strong bias dependence with the maximum occurring at a voltage drop well above 1 V. This special feature is accompanied by only a moderate temperature dependence of the tunnel magnetoresistance. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2711070]

I. INTRODUCTION

Co-based Heusler alloys are predicted to belong to the material class of the half metals.[1] They are characterized by a band gap at the Fermi energy for one spin direction and metallic properties for the other. Therefore, the charge carriers should be 100% spin polarized. Due to this feature large tunneling or giant magnetoresistance values are expected in layered structures incorporating Heusler alloys as one or both ferromagnetic electrodes.

In recent years, the Heusler compounds have been shown to yield increased tunneling magnetoresistance (TMR) values compared to the 3*d*-transition metals elements and their alloys. Still, the experimentally observed TMR values are way below the expectations. It is generally recognized that site disorder[2] and the presence of interface states[3] may drastically affect and even destroy the half metallicity. However, significant progress was recently made in improving the growth quality and reducing the site disorder. Thin films of Co-based Heusler alloys were successfully prepared in the ordered $L2_1$ structure.[4–6] These films display improved magnetic characteristics with Curie temperatures and magnetic moments close to the bulk values. Moreover, theoretical calculations[7] showed that Co-based full-Heusler alloys are more tolerant to site disorder, such that a high spin polarization is preserved even in the disordered *B*2 structure. Among the Co-based full-Heusler compounds, $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA) and Co_2MnSi (CMS) are promising candidates for obtaining large tunneling or giant magnetoresistance effects. They have large magnetic moments and high Curie temperatures (T_C), with CMS ($T_C \approx 985$ K) being among the highest.[8] Several groups have successfully prepared thin films of CCFA and CMS on various substrates, showing improved structural order and magnetic characteristics (see, for example, Refs. [4,6,9–11]). Remarkable progress was obtained also by incorporating these compounds into magnetic tunnel junctions (MTJs). Fully epitaxial junctions with Heusler CCFA films and MgO tunneling barriers showed 90% TMR ratio at room temperature (RT) and 240% at 4.2 K.[4] Presently, the largest TMR ratio for Heusler compounds is 570% at 2 K. It was obtained in layered structures comprising CMS as both ferromagnetic electrodes and an amorphous AlO_x barrier.[12]

We investigate thin films of CCFA and CMS full-Heusler compounds grown by magnetron sputtering at RT. In this contribution, we discuss the magnetic characteristics of CCFA and CMS thin films deposited directly onto oxidized Si wafers and report on large TMR values obtained at RT in layered structures combining a polycrystalline CCFA thin film with a MgO tunnel barrier and a CoFe counter electrode. The magnetoresistance shows on the one hand a rather moderate temperature dependence and on the other hand a strong bias dependence with the largest TMR value occurring at a high voltage drop clearly exceeding 1 V.

II. EXPERIMENT

CCFA and CMS thin films with a thickness of 25 nm as well as the MTJs were grown directly onto Si wafers covered with native, amorphous SiO_2 by magnetron sputtering at RT. After optimization, the sputtering Ar pressure was 1×10^{-3} mbar for CCFA films and 4.5×10^{-3} mbar for CMS films, respectively, at a target-substrate distance of 5 cm. We used stoichiometric $Co_2Cr_{0.6}Fe_{0.4}Al$ and Co_2MnSi targets for depositing our films. The sputtering rates, measured with a quartz balance, were calibrated using x-ray reflectivity. After deposition, the Heusler films can be annealed *in situ* up to 873 K. The structure of the Heusler films was identified *ex situ* by x-ray diffraction (XRD) carried out with a Philips X'Pert MRD diffractometer using Cu $K\alpha$ radiation. For the magnetic characterization, we employed superconducting quantum interference device (SQUID) magnetometry. Lay-

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ered structures are prepared without breaking the vacuum with the following layer sequence: SiO_2/CCFA (25 nm)/MgO (3 nm)/CoFe (5 nm)/IrMn (15 nm). The MgO barrier was deposited by rf-stimulated discharge from a stoichiometric target. Junctions with area from 3×3 up to 15×15 μm^2 with crossed electrodes were patterned for magnetotransport measurements in the current-perpendicular-plane (CPP) geometry by standard optical lithography. The transport measurements were performed with a dc setup in the four-point geometry using a constant current source. As for the SQUID measurements, the magnetic field is always applied in the film plane.

III. RESULTS AND DISCUSSION

Ideally, the full-Heusler compounds display the cubic, so-called $L2_1$ structure consisting of four interpenetrating fcc unit cells. Structural disorder occurs easily in this type of crystallographic structure. XRD (not shown) indicates the polycrystalline structure of our CCFA and CMS Heusler films deposited directly onto amorphous SiO_2 at RT. The CCFA films show a uniform distribution of grains with an average diameter of 40 nm and a rms roughness of 0.5 nm.[11] The films take the so-called $B2$ structure (CsCl type), where Co occupies the proper sublattice, while disorder occurs between the other elemental constituents. Postannealing has little influence on the crystallographic structure. More details about growth and structure characterization of CCFA films can be found in Ref. 11.

Despite the low deposition temperature and the polycrystalline growth, the magnetic characteristics of these films are comparable with results reported in literature for films deposited at higher temperature.[6,13,14] SQUID measurements of 25 nm thick CCFA and CMS films revealed ferromagnetic order with Curie temperatures well above RT. Annealing in vacuum at relatively high temperatures increases T_C for both CCFA and CMS films. The Curie temperature of the CCFA film can be increased up to 630 K after annealing in vacuum at 773 K for 1 h. In the case of CMS films, we performed annealing at 623 K for 1 h. The evaluation of the temperature dependence of the saturation magnetization based on SQUID data taken at temperatures up to 450 K indicates a T_C above 800 K. However, high temperature measurements are necessary to estimate the Curie temperature of CMS films more precisely. Both CCFA and CMS Heusler films display a soft ferromagnetic behavior. In Fig. 1, we plot the normalized M-H loops of 25 nm thick CCFA and CMS Heusler films, respectively, measured at RT. The coercive field H_c of the CCFA film is about 6 mT. Therefore, our CCFA films are magnetically softer compared to reported results about CCFA films deposited on Al_2O_3 substrates at high temperatures, which have a coercive field of 10 mT.[15] Moreover, the CMS film are magnetically even softer having a H_c of only 2 mT.

We estimated the total magnetic moments from the saturation magnetization at 5 K. As expected, the CMS films have higher magnetic moment than the CCFA layers. We find that annealing in vacuum increases not only T_C but also the value of the total magnetic moment. For the CCFA films we

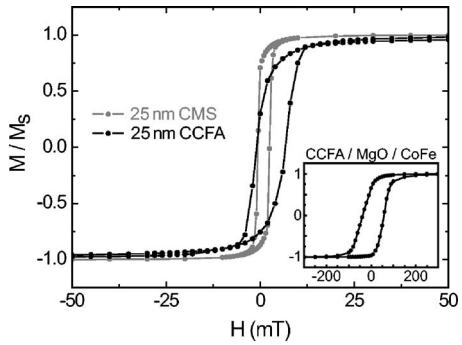

FIG. 1. Normalized magnetic hysteresis curves measured by SQUID at RT of 25 nm thick CCFA (black) and CMS (gray) films on SiO_2. The CCFA and CMS films are ferromagnetically soft with a H_c of 6 and 2 mT, respectively. The field biases are due to antiferromagnetic oxides that form on the surface of the uncapped films upon exposure to ambient air. Inset: magnetic hysteresis curve of a CCFA (25 nm)/MgO (3 nm)/CoFe (5 nm)/IrMn (15 nm) extended layered structure measured before lithography at 77 K.

obtained $2.56\mu_B$/f.u. and CMS films have a magnetic moment of $4.42\mu_B$/f.u. These values are lower than the predicted theoretical values, which are $3.8\mu_B$/f.u. for CCFA and $5\mu_B$/f.u. for CMS. These differences indicate the presence of site disorder in both materials. In the case of CCFA films, however, the value of the total magnetic moment is comparable with results obtained from films grown at elevated temperatures.[13] On the other hand, the CMS films have a total magnetic moment close to the experimental bulk value of $5.07\mu_B$/f.u..[8] We take this as an indication for less atomic disorder in the CMS films in comparison with the CCFA layers.

Magnetic tunneling junctions were prepared from stacks with the layer sequence SiO_2/CCFA (25 nm)/MgO (3 nm)/CoFe (5 nm)/IrMn (15 nm). The completed structure is annealed in vacuum for 1 h at 573 K. Although an IrMn antiferromagnet is located next to the upper CoFe electrode, the two ferromagnetic layers do not switch separately as observed in SQUID measurements performed on the extended multilayer before lithographically defining the MTJs (inset of Fig. 1). We mention at this point that no further treatment such as annealing above the Néel temperature of IrMn and field cooling could be applied to this structure. Thus, the CoFe electrode is not exchange biased. The bell-shaped magnetoresistance curves without separate switching fields are consistent with the SQUID measurements and indicate the presence of magnetic coupling between CCFA and CoFe. Antiferromagnetic, Néel-type and biquadratic couplings due to interface roughness are the most likely coupling mechanisms. This suggests a noncollinear orientation of the layer magnetizations in the field free state and, thus, the observed TMR values seem to be a lower limit. The switching behavior is different from that of fully epitaxial CCFA/MgO/CoFe MTJs with much lower roughness and thus negligibly small coupling reported by Marukame *et al.*[4]

In Fig. 2, we plot several magnetoresistance curves measured at RT with different bias currents I_{bias}. For this experiment, we used a constant current source and vary I_{bias} in steps of 0.1 mA. The TMR ratio is defined as $TMR=(R_{AP}-R_P)/R_P$, where R_{AP} and R_P are the resistances for the antiparallel and parallel magnetization configurations, respec-

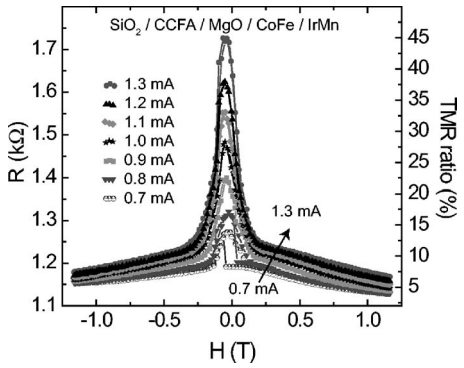

FIG. 2. Magnetoresistance curves of a 10×10 μm^2 junction with layer sequence SiO_2/CCFA (25 nm)/MgO (3 nm)/CoFe (5 nm)/IrMn (15 nm) measured by varying the bias current between 0.7 (bottommost curve) and 1.3 mA (topmost curve) in steps of 0.1 mA. The measurements are performed at RT and yield a maximum magnetoresistance of 46%. Joule heating at high currents might be the reason for the disappearing of hysteresis for currents larger than 0.9 mA.

tively. We obtained a maximum TMR value of 46% for $I_{bias}=1.3$ mA. For smaller I_{bias} the TMR ratio is much lower (about 1% for $I_{bias}\leqslant0.6$ mA) and also for larger I_{bias} the TMR ratio decreases, e.g., 18% for $I_{bias}=1.4$ mA and $<7\%$ for $I_{bias}=1.5$ mA. A variation of the bias current corresponds to a variation of the voltage drop across the junction. Multiplying the I_{bias} values in Fig. 2 by the corresponding resistance values in the saturated state, we obtain voltage drops in the range between 0.81 and 1.53 V. Therefore, the maximum TMR ratio occurs in these junctions at rather high voltages of more than 1 V. For all data presented here, the CCFA electrode is at positive voltage with respect to the CoFe counter electrode. The minimal resistance-area product is about 100 kΩ μm^2. The measured junction resistances exceed 1 kΩ in all samples and, thus, are much larger than the lead resistances ($\approx$10–20 Ω), indicating that there is no geometrical enhancement in the TMR ratios. The TMR values we obtained at RT are much larger compared to the results reported by Inomata *et al.*[10] in similar structures also comprising a polycrystalline CCFA electrode.

In Fig. 3, finally, we present the temperature dependence of the TMR effect. A weak temperature dependence is observed with the TMR ratio increasing only by a factor of about 2 from 30% at RT to 56% at 20 K. The measurements are performed with a constant bias current of 1 mA. A similarly weak temperature dependence was observed by Yamamoto *et al.*[16] in CCFA/MgO/CoFe structures, although the TMR values reported by these authors are slightly larger at low temperatures. The reason could be increased atomic order and better interfaces in their samples, which were prepared by molecular-beam epitaxy.

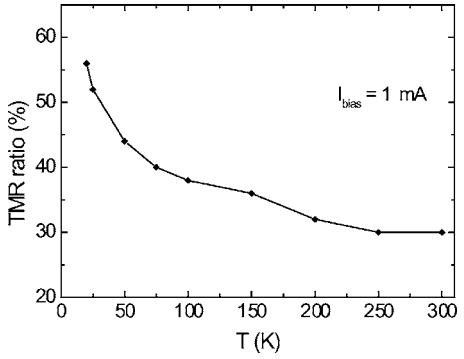

FIG. 3. Temperature dependence of the TMR ratio of a 10×10 μm^2 junction with layer sequence SiO_2/CCFA (25 nm)/MgO (3 nm)/CoFe (5 nm)/IrMn (15 nm). The measurements are done with a constant bias current of 1 mA.

IV. CONCLUSIONS

We prepared thin films of CCFA and CMS full-Heusler alloys by magnetron sputtering at room temperature. Our films are ferromagnetically soft, with coercive fields of about 6 mT for CCFA and 2 mT for CMS films. Annealing in vacuum increases the Curie temperature and the total magnetic moments. CCFA films combined with MgO barriers yield almost 50% TMR at room temperature. Most surprising, the high TMR ratios occur only for high voltage drops across the junctions of up to 1.5 V. The question whether this special magnetoresisitive behavior is related to spin filtering[17] at the MgO/CoFe interface of the "negative" electrode or to density-of-states effects in the "positive" CCFA electrode must be addressed in future experiments.

These results are encouraging for further investigations of TMR structures consisting of highly oriented full-Heusler alloy films combined with possibly epitaxial MgO tunneling barriers. From the application point of view, both the weak temperature dependence and the high voltage drop, which yields an output signal of 0.7 V at $I_{bias}=1.3$ mA, are of high relevance.

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