High tunnel magnetoresistance in fully epitaxial magnetic tunnel junctions with a full-Heusler alloy $Co_2Cr_{0.6}Fe_{0.4}Al$ thin film

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Fully epitaxial magnetic tunnel junctions (MTJs) were fabricated with a Co-based full-Heusler alloy $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA) thin film, whose composition was close to the stoichiometric one, and a MgO tunnel barrier. Cross-sectional high-resolution transmission electron microscope observations indicated that all layers of the CCFA/MgO/$Co_{50}Fe_{50}$ MTJ layer structure were grown epitaxially and were single crystalline. The microfabricated CCFA/MgO/$Co_{50}Fe_{50}$ MTJs exhibited high tunnel magnetoresistance (TMR) ratios of 90% at room temperature and 240% at 4.2 K. A high tunneling spin polarization of 0.79 at 4.2 K was obtained for the epitaxial CCFA films from the TMR ratios. [DOI: 10.1063/1.2217166]

In the field of spintronics, where spin-polarized electrons are employed, interest has been growing in the fabrication of magnetic tunnel junctions (MTJs) that use novel materials to achieve high tunnel magnetoresistance (TMR) ratios.[1,2] High TMR ratios of about 180% for single-crystal Fe(001)/MgO(001)/Fe(001) MTJs (Ref. 3) and about 220% for MTJs with a highly oriented MgO(001) tunnel barrier and $Co_{1-x}Fe_x$ electrodes[4] have been obtained at room temperature (RT). Subsequently, the TMR ratio was raised to 355% at RT through use of CoFeB electrodes and a highly oriented MgO(001) tunnel barrier.[5,6] Theoretical calculations suggest that high TMR ratios in fully epitaxial Fe/MgO/Fe MTJs result from the effective coupling, for the majority spin band, of one particular state with Δ_1 symmetry from the Fe into the MgO and also out of the MgO into the Fe electrode on the other side.[7]

Another approach is to develop MTJs using half-metallic ferromagnets (HMFs), which are characterized by an energy gap at the Fermi level (E_F) for the minority spin band, leading to complete spin polarization at E_F.[8] This characteristic provides the great advantage of enabling high TMR ratios in MTJs, according to Jullière's model.[9] Cobalt-based full-Heusler alloy thin films have been studied intensively because of the half-metallic nature theoretically predicted for some of these alloys[10,11] and because of their high Curie temperatures, which are well above RT.[12]

The Co-based full-Heusler alloy $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA) features high spin polarizations theoretically predicted for both the ordered $L2_1$ structure and the disordered $B2$ one,[13] along with a relatively high Curie temperature of 750 K.[14] The spin polarization theoretically predicted for the $L2_1$ structure is 0.90 and that for the $B2$ structure is 0.78.[13] The electronic and magnetic properties of the ordered and disordered full-Heusler alloys $Co_2Cr_{1-x}Fe_xAl$ with varying Cr to Fe ratio x have been studied theoretically and experimentaly.[15,16] Inomata *et al.* first demonstrated a relatively high TMR ratio of 16% at RT for MTJs using a Co-based full-Heusler alloy thin film—where the lower electrode was made of a polycrystalline CCFA thin film—and an amorphous AlO_x tunnel barrier.[17] Relatively high TMR ratios of up to 70% at RT have been reported for MTJs with an epitaxially grown Co-based full-Heusler alloy thin film (such as Co_2MnAl,[18] Co_2FeAl,[19] or Co_2MnSi[20]) as a lower electrode and an amorphous AlO_x tunnel barrier.

We recently reported fully epitaxial MTJs with a Co-based full-Heusler alloy thin film of either CCFA or Co_2MnGe as a lower electrode and a MgO tunnel barrier,[21–24] and obtained relatively high TMR ratios of 42% at RT and 74% at 55 K for epitaxial CCFA/MgO/$Co_{50}Fe_{50}$ MTJs.[21] However, much room remains for further enhancing the TMR ratio by preparing CCFA thin films having a composition close to the stoichiometric one of $Co_2(Cr_{0.6}Fe_{0.4})Al$, as well as by optimizing the MgO barrier thickness and fabrication conditions. One purpose of the present study was to investigate the TMR characteristics in fully epitaxial MTJs with a CCFA thin film having a composition close to the stoichiometric one, i.e., the 2:1:1 film composition of $Co_2(Cr_{0.6}Fe_{0.4})Al$. The second purpose was to deduce the effective spin polarization or the tunneling spin polarization P of the CCFA thin film from the obtained TMR ratios.

We fabricated fully epitaxial MTJs with a CCFA thin film having a composition close to the stoichiometric one and a wedge-shaped MgO tunnel barrier. The fabricated epitaxial MTJ layer structure (from the substrate side) was MgO buffer layer (10 nm)/CCFA lower electrode (50 nm)/MgO tunnel barrier (1.0–3.6 nm)/$Co_{50}Fe_{50}$ upper electrode (30 nm), and the structure was grown on a MgO(001) single-crystal substrate. Each layer in the MTJ layer structure was successively deposited in an ultrahigh vacuum chamber through the combined use of magnetron sputtering and electron beam evaporation. The CCFA layer was deposited at RT using magnetron sputtering and subsequently annealed *in situ* at 500 °C for 15 min. The MgO tunnel barrier was deposited at RT by electron beam evaporation. The $Co_{50}Fe_{50}$ layer, which had a coercive force higher than that of the CCFA layer, was deposited at RT using magnetron sputtering. The base pressure and the pressure during the deposition of the MgO tunnel barrier were 8×10^{-8} and 6×10^{-7} Pa, respectively. The fabrication procedure of the

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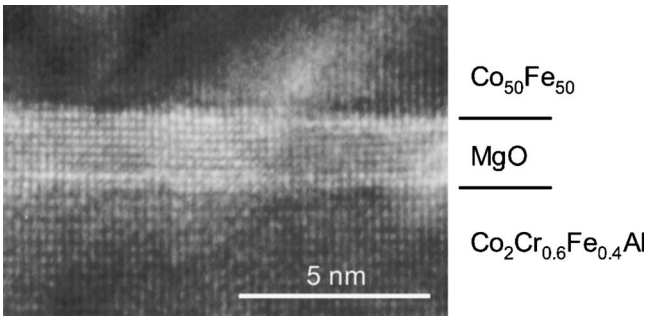

FIG. 1. Cross-sectional high-resolution transmission electron microscopy lattice image of a $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA)/MgO/$Co_{50}Fe_{50}$ MTJ layer structure along the [110] direction of CCFA. The nominal MgO thickness was 2.0 nm.

epitaxial MTJ layer structure has been described in detail elsewhere.[21–23] The composition of the fabricated CCFA film was $Co_{2.0}Cr_{0.56}Fe_{0.40}Al_{0.99}$, as determined through inductively coupled plasma analysis with an accuracy of 2%–3% for the composition of each element. Thus, the film composition was brought close to the stoichiometric one of 2:1:1 for $Co_2(Cr_{0.6}Fe_{0.4})Al$, in contrast with that of the $Co_{2.0}Cr_{0.61}Fe_{0.38}Al_{0.81}$ used to fabricate the CCFA-MTJs which showed TMR ratios of about 42% at RT.[23] The nominal thickness of the MgO tunnel barrier (t_{MgO}) was varied from 1.0 to 3.6 nm on each 20×20 mm^2 substrate by a linearly moving shutter during the deposition. We fabricated MTJs with the fully epitaxial layer structure by photolithography and Ar ion milling. The fabricated junction sizes were from 4×4 to 20×20 μm^2. After the microfabrication procedure, the MTJs were annealed at 175 °C for 1 h in a vacuum of 10^{-4} Pa under a magnetic field of 5 kOe. The magnetoresistance was measured with a magnetic field applied along the [110] axis of the CCFA at temperatures from 4.2 K to RT using a dc four-probe method. We defined the TMR ratio as $(RA_{AP}-RA_P)/RA_P$, where RA_{AP} and RA_P are the respective resistance-area products for the antiparallel and parallel magnetization configurations between the upper and lower electrodes.

Now we will describe the structural characterization results obtained from the fabricated CCFA films and MTJ layer structures. First, we confirmed that the fabricated CCFA films were epitaxial and crystallized in the *B*2 structure by x-ray pole figure measurements, which were in agreement with our previous work.[21–23] Microbeam electron diffraction patterns with beam diameters of 10–30 nm also indicated that the fabricated CCFA layer had the *B*2 structure. Figure 1 shows a cross-sectional high-resolution transmission electron microscope lattice image of a CCFA (50 nm)/MgO (2 nm)/$Co_{50}Fe_{50}$ (30 nm) MTJ layer structure along the [110] direction of the CCFA film. This image clearly shows that all the layers of the CCFA/MgO/$Co_{50}Fe_{50}$ MTJ structure were grown epitaxially and were single crystalline. It also confirmed that extremely smooth and abrupt interfaces were formed.

The as-fabricated (i.e., not *ex situ* annealed) MTJs exhibited typical TMR ratios of 80% at RT and 210% at 4.2 K. Figure 2 shows RA_P and TMR ratios of the as-fabricated MTJs at RT as a function of t_{MgO}. A clear exponential dependence of RA_P on t_{MgO} was observed for a t_{MgO} range from 1.1 to 2.5 nm, indicating typical tunnel junction behavior. High TMR ratios from 60% to 83% were obtained at RT for

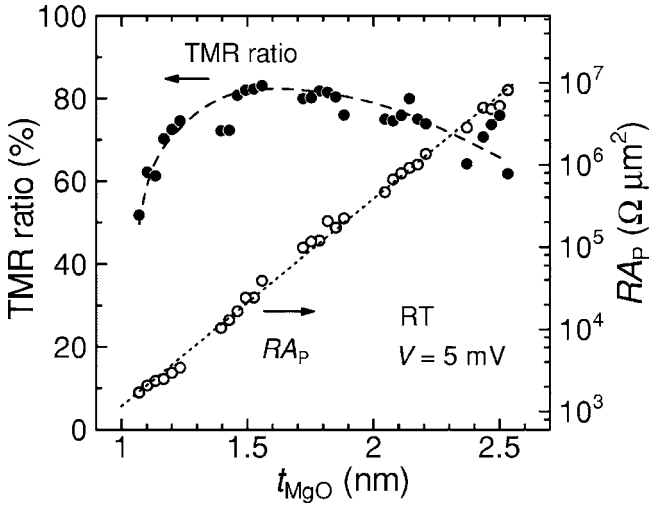

FIG. 2. TMR ratio and RA_P at RT (measured at a bias voltage of 5 mV) vs MgO tunnel barrier thickness t_{MgO} for as-fabricated (i.e., not *ex situ* annealed) $Co_2Cr_{0.6}Fe_{0.4}Al$/MgO/$Co_{50}Fe_{50}$ MTJs. RA_P represents the resistance-area product *RA* for the parallel magnetization configuration. The junction sizes were from 4×4 to 20×20 μm^2. The scale of the vertical axis for RA_P is logarithmic. The dashed line serves as a guide to the eye. The dotted line represents a least-squares approximation of the form $\ln(RA_P)=\alpha+\beta t_{MgO}$.

this wide range of t_{MgO} from 1.1 to 2.5 nm. Note that no significant dependence of the TMR ratio on t_{MgO} was observed over this range, which was similar to observations from epitaxial Fe/MgO/Fe MTJs (Ref. 3) and MTJs with a highly oriented MgO tunnel barrier and $Co_{1-x}Fe_x$ electrodes.[4] The significant increase of the TMR ratio in the epitaxial CCFA-MTJs, compared with our previously reported value of about 42% at RT,[21] suggests that a film composition close to the stoichiometric one is essential for obtaining high spin polarizations in CCFA thin films.

Figure 3 shows typical magnetoresistance curves at V =5 mV at RT and 4.2 K for a MTJ postfabrication annealed at 175 °C and having a 1.6-nm-thick MgO tunnel barrier.

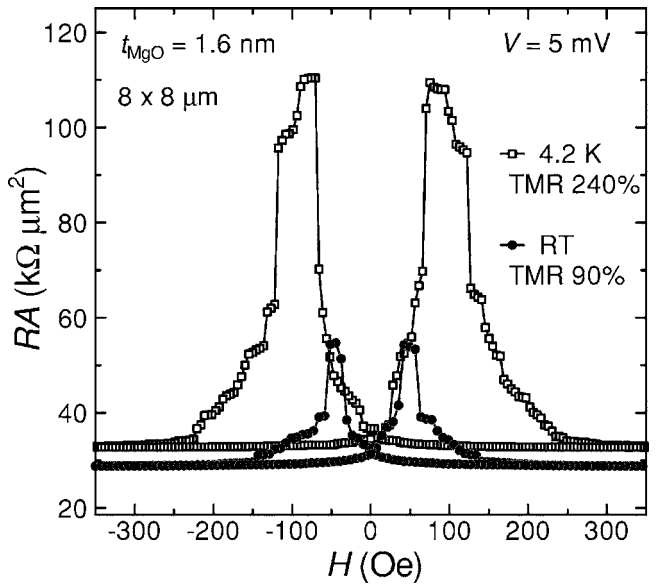

FIG. 3. Typical magnetoresistance curves for an epitaxial $Co_2Cr_{0.6}Fe_{0.4}Al$/MgO/$Co_{50}Fe_{50}$ MTJ *ex situ* annealed at 175 °C (t_{MgO} =1.6 nm) at a bias voltage of 5 mV at 4.2 K and RT. The junction size was 8×8 μm^2. The TMR ratios were 90% (RT) and 240% (4.2 K).

The annealed MTJs showed increased TMR ratios of 90% at RT and 240% at 4.2 K.

We deduced the spin polarization for the CCFA electrodes by using Jullière's model for the TMR ratio,[9] i.e., $\mathrm{TMR}=2P_1P_2/(1-P_1P_2)$, where P_1 and P_2 are the spin polarizations at E_F of the ferromagnetic electrodes in MTJs. The spin polarization P thus determined using Jullière's model with the TMR ratios of fully epitaxial, single-crystal MTJs (such as those used in this study) should be regarded as the effective spin polarization, or the tunneling spin polarization, rather than the originally defined spin polarization[9] with the density of states at E_F for the majority and minority spin bands. As a reference sample for the estimation, we fabricated fully epitaxial exchange-biased $Co_{50}Fe_{50}$/MgO/$Co_{50}Fe_{50}$ MTJs ($Co_{50}Fe_{50}$-MTJs). The layer structure (from the substrate side) was $Co_{50}Fe_{50}$ (50 nm)/MgO (0.8–3.4 nm)/$Co_{50}Fe_{50}$ (3 nm)/Ru (0.8 nm)/$Co_{90}Fe_{10}$ (2 nm)/IrMn (10 nm)/Ru (5 nm), and the structure was grown on a MgO-buffered MgO substrate. The $Co_{50}Fe_{50}$-MTJs were postfabrication annealed under the same annealing conditions as for the CCFA-MTJs (i.e., at 175 °C under a magnetic field of 5 kOe). The microfabricated $Co_{50}Fe_{50}$-MTJs showed typical TMR ratios of 125% at RT and 185% at 4.2 K. These TMR ratios for the epitaxial $Co_{50}Fe_{50}$-MTJs indicated that the tunneling spin polarization of the $Co_{50}Fe_{50}$ film (P_{CoFe}) was 0.69 at 4.2 K (0.62 at RT) according to Jullière's model. If we estimate the tunneling spin polarization of the CCFA film (P_{CCFA}) from the TMR ratio of 240% at 4.2 K (90% at RT) for the epitaxial CCFA-MTJs by using Jullière's model with P_{CoFe} of 0.69 at 4.2 K (0.62 at RT), we obtain a P_{CCFA} value of 0.79 at 4.2 K (0.50 at RT). This P_{CCFA} value of 0.79 at 4.2 K is close to the value of 0.78 theoretically predicted through the electronic band structure calculations for CCFA with the $B2$ structure.[13] The high tunneling spin polarization thus obtained confirms that fully epitaxial MTJs are promising as a key device structure for utilizing the potentially high spin polarizations of Co-based full-Heusler alloy thin films.

In summary, we obtained high TMR ratios of 90% at RT and 240% at 4.2 K in fully epitaxial $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA)/MgO/$Co_{50}Fe_{50}$ MTJs, where the CCFA thin film had a composition close to the stoichiometric one. A high tunneling spin polarization of 0.79 at 4.2 K was obtained for the epitaxial CCFA films from the TMR ratios.

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