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Highly Spin-Polarized Tunneling in Fully Epitaxial Magnetic Tunnel Junctions Using Full-Heusler Alloy $Co_2Cr_{0.6}Fe_{0.4}Al$ Thin Film and MgO Tunnel Barrier

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Highly spin-polarized tunneling with tunnel magnetoresistance (TMR) ratios of 90% at room temperature and 240% at 4.2 K was demonstrated for fully epitaxial magnetic tunnel junctions fabricated using a cobalt-based full-Heusler alloy $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA) thin film having a composition close to the stoichiometric one and a MgO tunnel barrier. A high tunneling spin polarization of 0.79 at 4.2 K was obtained for the epitaxial CCFA films from the TMR ratios. This adds to the promise of the fully epitaxial MTJ as a key device structure for utilizing the intrinsically high spin polarizations of Co-based full-Heusler alloy thin films.

***Index Terms*—Co-based full Heusler alloy, half-metallic ferromagnet, magnetic tunnel junction, tunnel magnetoresistance.**

I. INTRODUCTION

INTEREST has been growing in the fabrication of magnetic tunnel junctions (MTJs) with a half-metallic ferromagnet (HMF). An HMF is characterized by an energy gap for the minority-spin band, leading to a complete spin polarization at the Fermi level (E_F) [1]. The potentially high spin polarization of HMFs is advantageous for achieving high tunnel magnetoresistance (TMR) ratios in MTJs, efficient spin injection from ferromagnetic electrodes into semiconductors, and current-induced magnetization switching in MTJs. Cobalt-based full-Heusler alloy thin films have been studied intensively because of the half-metallic nature theoretically predicted for some of these alloys [2] and because of their high Curie temperatures [3], [4]. Inomata *et al.* first demonstrated a relatively high TMR ratio of 16% at room temperature (RT) for MTJs using a Co-based full-Heusler alloy thin film; these MTJs had a polycrystalline $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA) thin film as a lower electrode and an amorphous AlO_x tunnel barrier [5]. Recently, relatively high TMR ratios of up to 70% at RT have been shown for MTJs using a Co-based full-Heusler alloy thin film of Co_2MnSi and an amorphous AlO_x tunnel barrier [6].

We recently fabricated fully epitaxial MTJs using a Co-based full-Heusler alloy thin film of either $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA) or Co_2MnGe as a lower electrode and a MgO tunnel barrier [7]–[10] and obtained relatively high TMR ratios of 42% at RT and 74% at 55 K for CCFA/MgO/$Co_{50}Fe_{50}$ MTJs [7]. However, much room remains for further enhancing the TMR ratio by preparing CCFA thin films having a composition close to the stoichiometric one of $Co_2(Cr_{0.6}Fe_{0.4})Al$ and optimizing the MgO barrier thickness and other fabrication conditions. In this study, we fabricated fully epitaxial MTJs having a CCFA thin film whose composition was close to the stoichiometric one (i.e., the 2:1:1 film composition of $Co_2(Cr_{0.6}Fe_{0.4})Al$) and a wedge-shaped MgO tunnel barrier. We then investigated the TMR characteristics of these MTJs.

II. EXPERIMENTAL METHODS

The fabricated epitaxial MTJ layer structure was as follows: (from the substrate side) 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), 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 fabrication procedure of the epitaxial MTJ layer structure has been described in detail elsewhere [7]. The composition of the fabricated CCFA film was determined as $Co_{2.0}Cr_{0.56}Fe_{0.40}Al_{0.99}$ through inductively coupled plasma analysis with an accuracy of 2%–3% for the composition of each element. Thus, the film composition was certainly brought close to the stoichiometric one of 2:1:1 for $Co_2(Cr_{0.6}Fe_{0.4})Al$, and in contrast with that of $Co_{2.0}Cr_{0.61}Fe_{0.38}Al_{0.81}$, which was used to fabricate CCFA-MTJ's showing TMR ratios of about 42% at RT [9]. The nominal thickness of the MgO tunnel barrier (t_{MgO}) was varied from 1.0 to 3.6 nm on each 20×20 mm substrate by using a linearly moving shutter during the deposition. We fabricated MTJs with the fully epitaxial layer structure though photolithography and Ar ion milling. The fabricated junction sizes were from 4×4 μm to 20×20 μm. 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 $(RA_{\mathrm{AP}} - RA_{\mathrm{P}})/RA_{\mathrm{P}}$, where RA_{AP} and RA_{P} are

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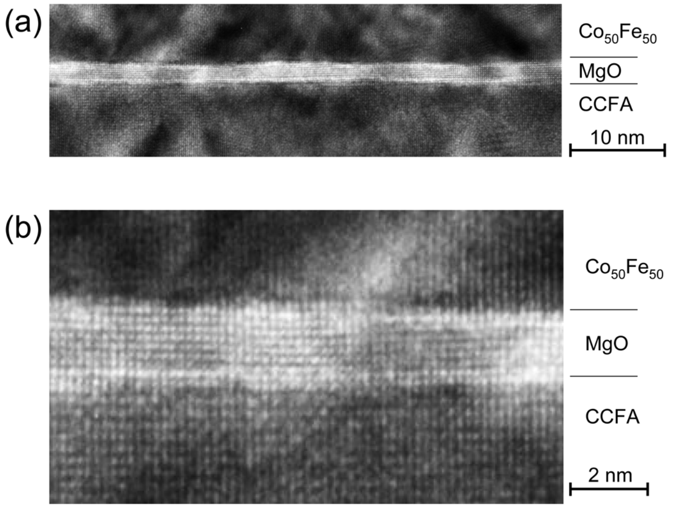

Fig. 1. (a) Cross-sectional high-resolution transmission electron microscopy lattice image of a $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA)/MgO/$Co_{50}Fe_{50}$ MTJ layer structure along the [110] direction of CCFA. The nominal MgO thickness was 2.0 nm. (b) A magnification of (a).

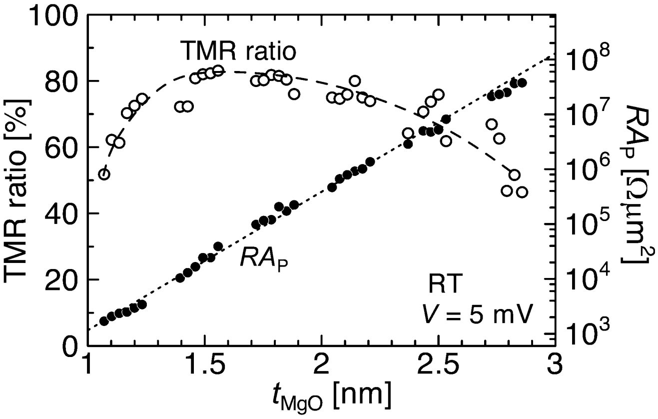

Fig. 2. TMR ratio and RA_P at RT (measured at a bias voltage of 5 mV) versus t_{MgO} for as-fabricated (i.e., not *ex situ* annealed) $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA)/MgO/$Co_{50}Fe_{50}$ MTJs. RA_P represents the RA product for the parallel magnetization configuration. The junction sizes were from 4 × 4 μm to 20 × 20 μm. 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 $\ln(RA_P) = \alpha + \beta \cdot t_{MgO}$.

the respective resistance-area products for the antiparallel and parallel magnetization configurations between the upper and lower electrodes.

III. EXPERIMENTAL RESULTS AND DISCUSSION

We investigated the structural properties of the fabricated CCFA films and MTJ layer structures. First, we confirmed that the fabricated CCFA films were epitaxial and crystallized in the B2 structure by X-ray pole figure measurements, which was in agreement with our previous work [7]–[9]. Microbeam electron diffraction patterns with beam diameters of 10–30 nm also indicated the fabricated CCFA layer had the B2 structure. Fig. 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 layer structure were grown epitaxially and were single-crystalline [Fig. 1(b)]. 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. The significant increase of the TMR ratio in the epitaxial CCFA-MTJs compared with our previously reported value of about 42% at RT [7] indicates that a film composition close to the stoichiometric one is essential to obtain high spin polarizations in CCFA thin films. Fig. 2 shows RA_P and TMR ratios of as-fabricated MTJs at RT as a function of t_{MgO}. As can be seen, a clear exponential dependence of RA_P on t_{MgO} was observed for a range of t_{MgO} from 1.1 to 2.8 nm, indicating typical tunnel junction behavior. The $m^*\phi_0$ value estimated from the slope of the $\ln(RA_P)$ versus t_{MgO} according to the Wenzel–Kramer–Brillouin (WKB) approximation was 0.32 eV for the CCFA-MTJ, where $m*$ is the effective electron mass normalized by the bare electron mass and ϕ_0 is the potential barrier height (the energy difference between the Fermi level of the emitter ferromagnetic electrode and the bottom of the conduction band in the tunnel barrier). This value was close to that of 0.39 eV for epitaxial Fe/MgO/Fe MTJs [11]. High TMR ratios from 50% to 83% were obtained at RT for a wide range of t_{MgO} from 1.1 to 2.8 nm. Note that no significant dependence of the TMR ratio on t_{MgO} over this wide range was similar to that observed for epitaxial Fe/MgO/Fe MTJs [11] and MTJs with a highly oriented MgO tunnel barrier and $Co_{1-x}Fe_x$ electrodes [12].

Fig. 3(a) shows typical magnetoresistance curves at $V = 5$ mV at RT and 4.2 K for an MTJ postfabrication annealed at 175 °C with a 1.6-nm-thick MgO tunnel barrier. Typical TMR ratios of 90% at RT and 240% at 4.2 K were obtained for the MTJ's postfabrication annealed at 175 °C.

One notable feature of the TMR characteristics is the strong temperature (T) dependence of the TMR ratio. Fig. 3(b) shows the TMR ratio and RA_{AP} and RA_P at a bias voltage of 5 mV as a function of T from 4.2 K to RT for a MTJ postfabrication annealed at 175 °C. With decreasing T from RT to 4.2 K, the TMR ratio increased 2.7 times from the value of 90% at RT to that of 240% at 4.2 K. As shown in Fig. 3(b), RA_{AP} also increased with decreasing T, while RA_P was almost independent of T. These results indicate that the increase in the TMR ratio with decreasing T was mainly due to the RA_{AP} increases. To clarify the physical origin of the observed marked temperature dependence of the TMR ratio of the fully epitaxial CCFA/MgO/CoFe MTJs, further detailed and systematic studies are needed.

We estimated the spin polarization for the CCFA electrodes by using Jullière's model for the TMR ratio; i.e., TMR ratio $= 2P_1P_2/(1 - P_1P_2)$, where P_1 and P_2 are the spin polarizations at E_F of the ferromagnetic electrodes in MTJs [13]. Thus, the spin polarization estimated using Jullière's model for fully epitaxial, single-crystal MTJs, such as those used in this study, should be regarded as an effective spin polarization, or a tunneling spin polarization. As a reference sample for the estimation, we also fabricated fully epitaxial exchange-biased $Co_{50}Fe_{50}$/MgO/$Co_{50}Fe_{50}$ MTJs ($Co_{50}Fe_{50}$-MTJs). The layer

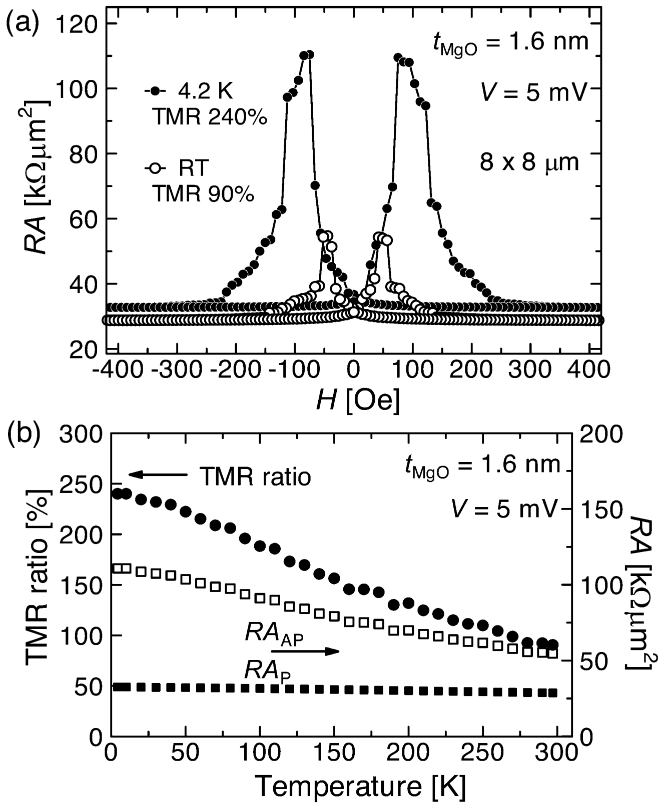

Fig. 3. (a) Typical magnetoresistance curves for an epitaxial $Co_2Cr_{0.6}Fe_{0.4}Al/MgO/Co_{50}Fe_{50}$ MTJ ($t_{MgO} = 1.6$ nm) at a bias voltage of 5 mV at 4.2 K and RT. The junction size was 8 × 8 μm. The TMR ratios were 90% (RT) and 240% (4.2 K). (b) TMR ratios at a bias voltage of 5 mV and RA_{AP} and RA_P for the MTJ as a function of temperature from 4.2 K to RT, where RA_{AP} and RA_P represent junction resistance-area products for, respectively, the antiparallel and parallel magnetization configurations.

structure was (from the substrate side) $Co_{50}Fe_{50}$ (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), grown on a MgO-buffered MgO substrate. The $Co_{50}Fe_{50}$-MTJs were annealed under the same 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. Using these obtained TMR ratios with Jullière's model resulted in an effective spin polarization for the $Co_{50}Fe_{50}$ film (P_{CoFe}) of 0.69 at 4.2 K (0.62 at RT). If we estimate the effective spin polarization of the CCFA film (P_{CCFA}) from the TMR ratio of 240% at 4.2 K (90% at RT) by using Jullière's model with the 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 (0.79 at 4.2 K) was close to the value of 0.78 theoretically predicted through the band structure calculation for CCFA with the B2 structure [14]. The obtained high spin polarization supports the promise of fully epitaxial MTJ layer structures as a key device structure for utilizing the intrinsically high spin polarization of Co-based full-Heusler alloy thin films.

IV. Summary

Highly spin-polarized tunneling was demonstrated for fully epitaxial magnetic tunnel junctions (MTJs) fabricated using a cobalt-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. The microfabricated MTJs exhibited high tunnel magnetoresistance ratios of 90% at RT and 240% at 4.2 K. A high effective spin polarization of 0.79 at 4.2 K was obtained for the epitaxial CCFA films.

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