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Tunnel magnetoresistance in epitaxial magnetic tunnel junctions using full-Heusler alloy Co_2MnGe thin film and MgO tunnel barrier

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We fabricated fully epitaxial magnetic tunnel junctions (MTJs) using a Co-based full-Heusler alloy Co_2MnGe (CMG) thin film as a lower electrode, an MgO tunnel barrier, and a $Co_{50}Fe_{50}$ upper electrode and investigated their tunnel magnetoresistance (TMR) characteristics. The microfabricated MTJs showed strongly temperature-dependent TMR characteristics with typical TMR ratios of 70% at 7 K and 14% at room temperature (RT). Furthermore, the TMR characteristics exhibited the following notable features in the bias voltage (V) dependence: (1) a cusplike V dependence within a range of $\sim\pm 200$ mV around $V=0$ at 7 K, which was smeared out at RT, and (2) a V dependence with pronounced asymmetry regarding the polarity, showing negative TMR ratios in a certain negative bias voltage range around -400 mV at both 7 K and RT (V was defined with respect to the lower CMG electrode). A possible transport mechanism leading to the notably asymmetric V dependence along with the negative TMR for a certain bias voltage region is direct tunneling that reflects the spin-dependent density of states of the CMG electrode. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2167063]

Cobalt-based full-Heusler alloy thin films have recently been studied extensively as highly preferable ferromagnetic electrodes for spintronic devices.[1–7] This is because some Co-based full-Heusler alloys have been theoretically predicted to be half-metallic ferromagnets[8–10] (HMFs) and because of their high Curie temperatures, which are well above room temperature (RT).[11] HMFs 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,[12] which is highly desirable for spintronic applications. Recently, relatively high tunnel magnetoresistance (TMR) ratios have been reported for magnetic tunnel junctions (MTJs) using Co-based full-Heusler alloys, including $Co_2Cr_{0.6}Fe_{0.4}Al$ (CCFA),[3,6] Co_2MnSi (CMS),[4] and Co–Mn–Al.[5] Moreover, strong temperature and bias voltage dependences in MTJs using a polycrystalline Co_2MnSi thin film as a lower electrode and an amorphous AlO_x tunnel barrier (CMS-MTJ) have been reported, and the inelastic tunneling via an electron-magnon interaction has been considered a possible transport mechanism for these dependences.[4,13]

Co_2MnGe (CMG) is one of the Co-based full-Heusler alloys that has been theoretically predicted to be half-metallic[8,9] with a minority-spin band energy gap of $\sim$0.21 eV.[8] Up to now, epitaxial $L2_1$-structured Co_2MnGe thin films have been grown on GaAs substrates using molecular-beam epitaxy[1,7] (MBE) or pulsed laser deposition,[2] and spin injection from CMG into compound semiconductor heterostructures has been studied.[7] However, MTJs using a CMG thin film have not yet been developed. We recently reported fully epitaxial MTJs using a CCFA thin film as a lower electrode and an MgO tunnel barrier and demonstrated relatively high TMR ratios of 42% at RT and 74% at 55 K.[6]

In our current study, we fabricated fully epitaxial MTJs using a CMG thin film as a lower electrode and an MgO tunnel barrier and investigated their TMR characteristics as a function of temperature (T) and bias voltage (V).

The MTJ layer structure was made by successively depositing (from the substrate side) an MgO buffer layer (10 nm), a CMG lower electrode (50 nm), an MgO tunnel barrier (2.5–3.0 nm), and a $Co_{50}Fe_{50}$ (CoFe) upper electrode (17 nm) on an MgO(001) single-crystal substrate. The CMG layer was deposited at RT using magnetron sputtering and subsequently annealed *in situ* at 600 °C for 15 min.[14] The MgO tunnel barrier was deposited at RT by electron-beam evaporation. The CoFe layer, which had a coercive force (H_c) higher than that of the CMG layer, was deposited at RT using magnetron sputtering to form parallel and antiparallel magnetization configurations between the lower and upper electrodes. The composition of the fabricated CMG film was determined as $Co_{2.00}Mn_{1.05}Ge_{1.17}$ through inductively coupled plasma analysis with an accuracy of 2%–3% for the composition of each element. X-ray-diffraction pole figure measurements indicated that the CMG grew epitaxially and crystallized in the $L2_1$ crystal structure. Furthermore, microbeam electron diffraction patterns with beam diameters of 10–30 nm indicated the $L2_1$ structure for a fabricated film annealed at 600 °C as well as some residual regions of the B2 and A2 structures. Detailed structural and magnetic properties of the CMG thin films will be reported elsewhere.[14]

We fabricated MTJs with the fully epitaxial layer structure by photolithography and Ar-ion milling. The junction sizes were from 3×3 to 10×10 μm^2. The magnetoresistance was measured with the magnetic field applied along the

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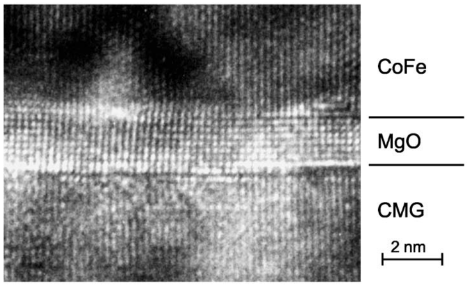

FIG. 1. Cross-sectional HRTEM lattice image of a $Co_2MnGe/MgO/CoFe$ heterostructure along the [1-10] direction of CMG.

[110] axis of the CMG within a temperature range from 7 K to RT using a dc four-probe method. The differential conductance $G(=dI/dV)$ was measured using standard ac lock-in techniques. We defined the TMR ratio as $(R_{AP}-R_P)/R_P$, where R_{AP} and R_P are the respective resistances for the antiparallel and parallel magnetization configurations between the upper and lower electrodes. We also defined the differential TMR ratio as $(G_P-G_{AP})/G_{AP}$, where G_P and G_{AP} represent G for the parallel and antiparallel magnetization configurations, respectively.

Now we will describe the experimental results obtained from the fabricated MTJs. Figure 1 shows a cross-sectional high-resolution transmission electron microscopy (HRTEM) lattice image of a CMG/MgO/CoFe heterostructure along the [1-10] direction of the CMG. It clearly shows that all layers of the CMG/MgO/CoFe heterostructure were grown epitaxially and single crystalline. Furthermore, it confirmed that extremely smooth interfaces were obtained.

Typical magnetoresistance curves for a fabricated MTJ with a 3.0-nm-thick MgO tunnel barrier at $V=+5$ mV (the bias voltage was defined with respect to the lower CMG electrode) at RT and 7 K are shown in Fig. 2(a). The fabricated MTJs showed clear TMR characteristics with typical TMR ratios, at a small positive V of 5 mV, of 70% at 7 K and 14% at RT. The resistance and junction area product RA for MTJs with a 3.0-nm-thick MgO tunnel barrier was typically 10 $k\Omega\,\mu m^2$ at RT [Fig. 2(a)]. The first notable feature of their TMR characteristics is the strong temperature dependence of the TMR ratio at a small positive V of 5 mV [Fig. 2(b)]. For comparison, the TMR ratio as a function of T is also plotted for a fully epitaxial CCFA/MgO/CoFe MTJ (reference CCFA-MTJ) identically fabricated except that the lower electrode CMG was replaced with CCFA.[6] With decreasing T from RT to 7 K, the TMR ratio of the CMG-MTJ increased fivefold, while that of the reference CCFA-MTJ increased by no more than about 80%. In addition, the relative increase of the TMR ratio of the CMG-MTJ with decreasing T from RT to 7 K was twice that of a polycrystalline CMS-MTJ.[4]

Figure 3 shows the TMR ratio as a function of V at RT and 7 K. As shown, the TMR characteristics exhibited the following notable features in the bias voltage dependence: (1) a cusplike V dependence within a range of ~±200 mV around $V=0$ at 7 K, which was smeared out at RT, and (2) a V dependence with pronounced asymmetry regarding the polarity, showing negative TMR ratios in a certain negative bias voltage range around −400 mV at both 7 K and RT.

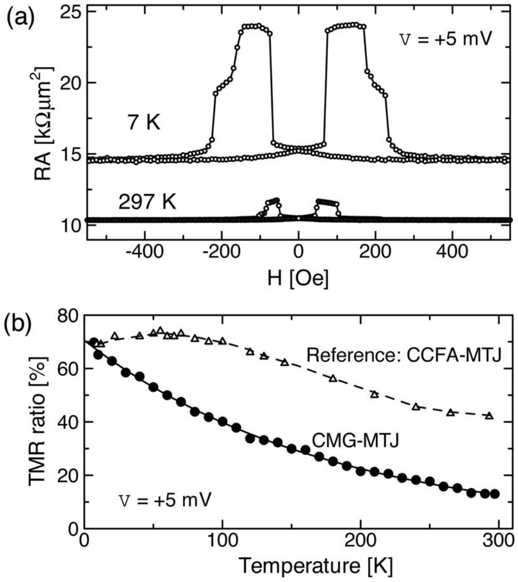

FIG. 2. (a) Typical magnetoresistance curves for an epitaxial $Co_2MnGe/MgO/CoFe$ MTJ (CMG-MTJ) at bias voltage V of +5 mV at 7 and 297 K. The TMR ratios were 14% (297 K) and 70% (7 K). (b) Temperature dependence of the TMR ratio of the CMG-MTJ at $V=+5$ mV in comparison with that of the reference MTJ consisting of an identically fabricated epitaxial $Co_2Cr_{0.6}Fe_{0.4}Al/MgO/CoFe$ layer structure (reference CCFA-MTJ). The solid and dashed lines serve as a guide to the eye.

The strong T dependence and the cusplike V dependence of the TMR ratio observed for the CMG-MTJs are like those observed for CMS-MTJs.[4,13] These characteristics for the latter have been discussed in terms of magnon-assisted inelastic tunneling.[13,15] The most notable feature in the TMR characteristics of the CMG-MTJs is, however, the highly asymmetric bias voltage dependence which shows negative TMR ratios in a certain negative bias voltage range around −400 mV. Inelastic tunneling via, for example, electron-magnon interaction can hardly explain the observed bias-voltage-dependent negative TMR ratios. However, a direct tunneling mechanism that reflects the spin-dependent density of states of ferromagnetic electrodes is a candidate to explain the observed bias-voltage-dependent negative TMR ratios.

To further investigate spin-dependent magnetotransport properties in the CMG-MTJs, we measured the bias voltage dependence of the differential conductance $(G=dI/dV)$ at RT

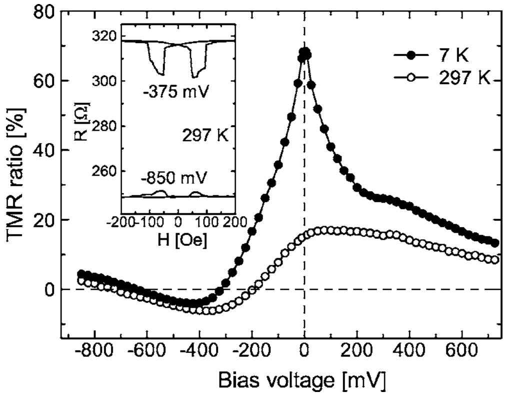

FIG. 3. Bias voltage dependence of the TMR ratio at 7 K and RT for an epitaxial $Co_2MnGe/MgO/CoFe$ MTJ. The inset shows the magnetoresistance curves at RT, showing a negative TMR ratio at $V=-375$ mV and a positive TMR ratio at $V=-850$ mV.

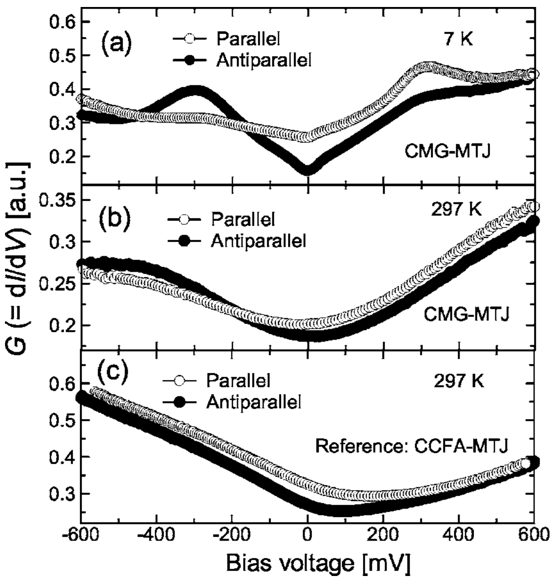

FIG. 4. Bias voltage dependence of the differential conductances G_P and G_{AP} of an epitaxial $Co_2MnGe/MgO/CoFe$ MTJ (CMG-MTJ), where G_P and G_{AP} represent $G=dI/dV$ for the parallel and antiparallel magnetization configurations, respectively. (a) CMG-MTJ at 7 K, (b) CMG-MTJ at 297 K, and (c) $Co_2Cr_{0.6}Fe_{0.4}Al/MgO/CoFe$ MTJ (reference CCFA-MTJ) at 297 K for comparison.

and 7 K. Figure 4 shows $G_P(V)$ and $G_{AP}(V)$ at RT and 7 K along with those for the reference CCFA-MTJ at RT. Figure 4 shows, although G_P was larger than G_{AP} over the positive bias voltage region for the CMG-MTJ at both RT and 7 K, a clear crossover between G_P and G_{AP} occurred with increasing negative bias voltage. This crossover means negative differential TMR characteristics for a certain negative bias voltage region. These negative differential TMR characteristics correspond to the negative TMR characteristics observed for a certain negative region of V. In contrast, the measured $G_P(V)$ and $G_{AP}(V)$ at RT for the reference CCFA-MTJ showed that G_P was always larger than G_{AP} over both positive and negative bias regions. Furthermore, the measured $G_P(V)$ and $G_{AP}(V)$ at RT for a reference MTJ with an identically fabricated epitaxial CoFe/MgO/CoFe heterostructure (CoFe-MTJ) also showed that G_P was always larger than G_{AP} over both positive and negative bias regions. These comparisons suggest that the observed negative TMR characteristics for the CMG-MTJ were due to direct tunneling which reflected the bulk spin-dependent electronic density of states rather than that related to interface states[16] and were due to the spin-dependent density of states of the CMG electrode rather than that of the CoFe electrode. Note also that the increase of G_{AP} near $V\sim -300$ mV for the CMG-MTJ was significantly enhanced at 7 K, and the characteristic increase of G_P appeared near $V\sim +300$ mV. These features can hardly be explained by inelastic tunneling. The direct tunneling mechanism that reflects the spin-dependent density of states of the ferromagnetic electrodes can also be a dominant factor accounting for the characteristic structures in the $G_{AP}(V)$ and $G_P(V)$ observed at 7 K. For the negative V and the antiparallel magnetization configuration, electrons, being just below E_F, of the majority-spin band of the CoFe electrode tunnel into the electronic states, being above eV from E_F, of the minority-spin band of the CMG one. On the other hand, for the positive V and the parallel magnetization configuration, electrons, being below eV from E_F, of the minority-spin band of the CMG electrode tunnel into the electronic states, being just above E_F, of the minority-spin band of the CoFe one. If we assume the CMG electrode's spin-dependent density of states is dominant rather than the CoFe one, the characteristic increase of G_{AP} appeared near $V\sim -300$ mV indicates the existence of the peak around 0.3 eV above E_F in the minority-spin band density of states of the CMG, while the characteristic increase of G_P appeared near $V\sim +300$ mV indicates the existence of the peak around −0.3 eV below E_F in the minority-spin band density of states of the CMG. Thus, a full analysis of the $G_P(V)$ and $G_{AP}(V)$ characteristics will provide information on the spin-dependent electronic structure of the epitaxial CMG electrode.

In summary, we fabricated fully epitaxial MTJs using a Co-based full-Heusler alloy—Co_2MnGe (CMG)—thin film as a lower electrode and an MgO tunnel barrier and investigated their tunnel magnetoresistance (TMR) characteristics. The microfabricated MTJs exhibited strongly temperature-dependent TMR characteristics with typical TMR ratios of 70% at 7 K and 14% at RT. Furthermore, the TMR characteristics exhibited bias voltage (V) dependence that was highly asymmetric with respect to the polarity of V, showing negative TMR in a certain negative bias voltage range around −400 mV at both 7 K and RT (V was defined with respect to the lower CMG electrode). A transport mechanism that could lead to this notably asymmetric V dependence along with the negative TMR for a certain bias voltage region is direct tunneling that reflects the spin-dependent density of states of the CMG electrode.

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