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Enhancement of current-perpendicular-to-plane giant magnetoresistive outputs by improving B2-order in polycrystalline $Co_2(Mn_{0.6}Fe_{0.4})Ge$ Heusler alloy films with the insertion of amorphous CoFeBTa underlayer

S. Li [a, 1], T. Nakatani [a, *], K. Masuda [a], Y. Sakuraba [a], X.D. Xu [a], T.T. Sasaki [a], H. Tajiri [b], Y. Miura [a, c], T. Furubayashi [a], K. Hono [a]

[a] National Institute for Materials Science, 1-2-1 Sengen, Tsukuba, Ibaraki, 305-0047, Japan
[b] Japan Synchrotron Radiation Research Institute/SPring-8, 1-1-1, Sayo, Hyogo, 679-5198, Japan
[c] Kyoto Institute of Technology, Matsugasaki, Sakyo-ku, Kyoto, 606-8585, Japan

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ABSTRACT

We studied the origin of the enhancement of current-perpendicular-to-plane giant magnetoresistance (CPP-GMR) effect by inserting a thin amorphous CoFeBTa (CFBT) underlayer below a $Co_2(Mn_{0.6}Fe_{0.4})Ge$ (CMFG) Heusler alloy ferromagnetic (FM) layer. Large magnetoresistance ratio of ~25% and resistance change-area product of ~7.5 mΩ μm^2 were obtained at room temperature by inserting a CFBT (1.2 nm) underlayer. X-ray diffraction (XRD) and transmission electron microscope analyses confirmed that the CMFG FM layer deposited on the CFBT underlayer was amorphous in the as-deposited state and crystallized to a B2-ordered polycrystalline film by annealing at 300 °C. The degree of B2 order (S_{B2}) of the CMFG films was estimated by anomalous XRD using x-ray energies around the Co *K*-absorption edge. S_{B2} of the CMFG film deposited on the amorphous CFBT (1.2 nm) underlayer was ~0.76, much larger than that of the CMFG film deposited on a crystalline CoFe underlayer (S_{B2} ~0.47). First-principles calculations indicated that the spin polarization of the *sp*-conduction electrons in CMFG increases with increasing S_{B2}, which accounts for the enhanced CPP-GMR effect in the pseudo spin-valve by inserting an amorphous CFBT underlayer.

1. Introduction

Due to the theoretically predicted half-metallicity and high Curie temperature, Co_2YZ (Y = Mn and Fe; Z = Si, Ge, Al, and Ga *etc.*) full-Heusler alloys [1] have received extensive research interest for spintronic applications such as tunnel magnetoresistance (TMR) [2,3], current-perpendicular-to-plane giant magnetoresistance (CPP-GMR) [4–7], lateral spin-valves [8,9] and spin injectors into semiconductors [10,11]. Especially, the large magnetoresistive (MR) outputs of CPP-GMR sensors using Heusler alloy ferromagnetic (FM) layers are promising for the read head sensors of ultrahigh recording density hard disk drives (HDDs) [12–14]. This is because of the low device resistance of all-metallic CPP-GMR films, which is beneficial for a low-noise operation of the read sensors with small dimensions (<30 nm). So far, large MR values have been reported for the CPP-GMR devices using epitaxially grown single-crystalline Heusler alloy films annealed at high temperatures (≥500 °C) [15,16]. Such a high temperature annealing enables a high degree of $L2_1$ or B2 order in the Heusler alloys, by which a high spin polarization of conduction electrons can be obtained. For real device applications, however, large MR values must be realized with a realistic device structure fabricated under various restrictions of the process conditions. Specifically, the read sensors of HDDs must have the spin-valve structure deposited on electrodeposited polycrystalline NiFe shield; hence, the read sensor films are expected to be polycrystalline. The layer structure of the thin polycrystalline spin-valve films is destroyed at elevated temperature due to the recrystallization of the polycrystalline films. For example, the

* Corresponding author.
E-mail address: nakatani.tomoya@nims.go.jp (T. Nakatani).
[1] Present address: Takasaki Advanced Radiation Research Institute, National Institutes for Quantum and Radiological Science and Technology, 1233 Watanuki, Takasaki, 370-1292, Japan.

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magnetic and MR properties of the current spin-valve read sensors are degraded at above 300–400 °C [17]. Therefore, realizing a high degree of order in polycrystalline Heusler alloy films at a relatively low annealing temperature (~300 °C) is the key to realize CPP-GMR read sensors.

Very recently, Choi et al. reported that the MR ratio of polycrystalline CPP-GMR sensors using $Co_2(Mn_{0.6}Fe_{0.4})Ge$ (CMFG) Heusler alloy FM layer could be improved by inserting a thin CoFeBTa (CFBT) amorphous layer below the CMFG pinned layer [18], in which ~7 at. % of Ta was added to CoFeB to stabilize the amorphous structure up to 450 °C. They attributed the enhancement of CPP-GMR by the insertion of CFBT to an improvement of degree of B2 order of the CMFG film based on x-ray diffraction (XRD) measurements by a laboratory XRD tool with the Cu K_α radiation source. The B2 order is a chemical order between Co and *YZ*, *i.e.* the chemical order between Co and (Mn, Fe, Ge) in the case of CMFG. Generally, the degrees of $L2_1$ and B2 order have a significant influence on the spin polarization of Heuser alloys. Picozzi et al. [19] and Miura et al. [20] have shown that at least B2 order is needed for the half-metallic band structure of Co_2YZ Heusler alloys, and that the spin polarization decreases rapidly with increasing Co-*Y* disorder (Co-(Mn, Fe) disorder in the case of CMFG). However, since the atomic scattering factors of those 3*d* elements for laboratory x-ray sources such as the Cu K_α radiation are close to each other, the chemical order between Co and *Y* is not measurable. Thus, the B2 superlattice diffraction peak measured by a laboratory XRD tool in the work by Choi et al. [18] indicates only the chemical order between Co and *Z* (Ge) while the intensity of the B2 superlattice diffraction peak is independent of the degree of Co-*Y* order. This means that the "B2 structure" measured by the laboratory XRD tool can be actually a $D0_3$ (Co, *Y*)$_3$Z structure where Co and *Y* are completely disordered [21,22]. Therefore, quantitative characterization of the chemical order between Co and *Y* sites is crucial to correlate the degree of order of Heusler alloy films and their magnetoresistive properties. An effective way to characterize the chemical order between the 3*d* elements is the XRD measurement utilizing an anomalous dispersion effect. In the case of Co-based Heusler alloys, the atomic scattering factor of Co is modulated near the *K*-absorption edge of Co at 7709 eV of the x-ray energy, so that the intensity of B2 superlattice diffraction changes depending on the degree of order between Co and (Mn, Fe). By measuring the intensities of B2 peak at various x-ray energies around the Co-*K* absorption edge, the degree of B2 order can be estimated [22,23].

In this study, we carried out a comprehensive experimental investigation on the influence of the CFBT underlayer on the degree of B2 order of CMFG films and on the CPP-GMR output. The relationship between the degree of B2 order of CMFG and the spin polarization was also studied by first-principles calculations. We found that the degree of B2 order of the polycrystalline CMFG film was significantly enhanced by inserting a CFBT underlayer below the CMFG layer, by which the CPP-GMR output was increased. The first-principles calculations showed that the spin polarization of the conduction electron in CMFG increased with increasing degree of B2 order. Thus, the enhanced CPP-GMR output by the insertion of CFBT is concluded to be due to the improved B2 order of the CMFG layer.

2. Methods

2.1. First-principles calculations

First-principles calculations were carried out using the AkaiKKR code [24] based on the Korringa-Kohn-Rostoker method in combination with the density-functional theory [25,26]. For the exchange-correlation energy, we adopted the local density approximation with the parametrization given by Moruzzi, Janak and Williams [27,28]. We also used the coherent potential approximation to treat the chemical disorder in CMFG, where the space was divided into non-overlapping muffin-tin spheres. In our calculations, the spin-orbit interactions were neglected and 4096 (=16 × 16 × 16) **k** points were used for the Brillouin-zone integrations. As parameters, we chose the imaginary part of the energy as 0.001 Ry and the width of the energy contour as 1.5 Ry.

In the $L2_1$-ordered Heusler alloys X_2YZ, (0,0,0), (1/2,1/2,1/2), (1/4,1/4,1/4), and (3/4,3/4,3/4) sites in Wyckoff coordinates are occupied by *X*, *X*, *Y*, and *Z* atoms, respectively (Fig. 1(a)), where *X*, *Y*, and *Z* correspond to Co, $(Mn_{0.6}Fe_{0.4})$, and Ge in the case of CMFG. If *Y* and *Z* sites are completely disordered, the structure becomes a B2 structure expressed explicitly by *XY*, as shown in Fig. 1(b). In the present work, we additionally considered chemical disorders between Co and *Y* (=Mn, Fe, and Ge) sites in the B2-ordered CMFG, $Co_2(Mn_{0.3}Fe_{0.2}Ge_{0.5})_2$, where we define the amount of disorder *x* by the ratio of the Co sites occupied by *Y* atoms. In this paper, we call *x* "ratio of Co-site disorder". For example, when we consider the disorder between Co and Fe atoms, the chemical formula is given by $(Co_{1-x}Fe_x)_2(Mn_{0.3}Fe_{0.2-x}Co_xGe_{0.5})_2$. Using *x*, the degree of B2 order (S_{B2}) is defined by $S_{B2} = 1-2x$. Following this definition, $S_{B2} = 0$ and 1 correspond to the completely disordered A2 structure and the perfectly ordered B2 structure, respectively. For all the calculations, we used the lattice parameter $a = 0.5743$ nm expected for CMFG with the $L2_1$ structure by applying the Vegard's law to the identical lattice parameters of the terminal alloys, Co_2MnGe [29] and Co_2FeGe [30], because the lattice parameter of the bulk CMFG has not been reported experimentally so far.

2.2. CPP-GMR film and device fabrications

Polycrystalline CPP-GMR thin films were deposited by ultrahigh vacuum magnetron sputtering at room temperature. CMFG Heusler alloy [31] films were deposited by dc sputtering from a sintered Co-Mn-Fe-Ge alloy target. The composition of the CMFG film was measured by x-ray fluorescence to be $Co_{51.0}Mn_{14.4}Fe_{9.8}Ge_{24.8}$ (at. %). Pseudo spin-valve (PSV) structure of Cu(100)/Ta-based bottom lead/Ru(2)/CoFe(1)/CFBT(0–1.2)/CMFG(5)/CoFe(0.4)/AgSn(4)/CoFe(0.4)/CMFG(5)/CoFe(1)/Ru(8) cap (thickness in nanometer) were deposited. The nominal compositions of AgSn, CoFe and CFBT were $Ag_{90}Sn_{10}$, $Co_{50}Fe_{50}$ and $Co_{37}Fe_{37}B_{19}Ta_7$ (at. %), respectively. The AgSn alloy was used for the spacer layer due to the nature of flat growth of the polycrystalline AgSn film, which is important for the spacer layer of CPP-GMR device [32]. After the completion of the sputtering deposition, the PSV stacks were annealed at 300 °C for 3 h under a magnetic field of 0.3 T, then patterned to elliptical shaped pillars of an approximately 150 nm × 300 nm dimension, encapsulated by SiO_2, and a Cu top contact was formed for CPP

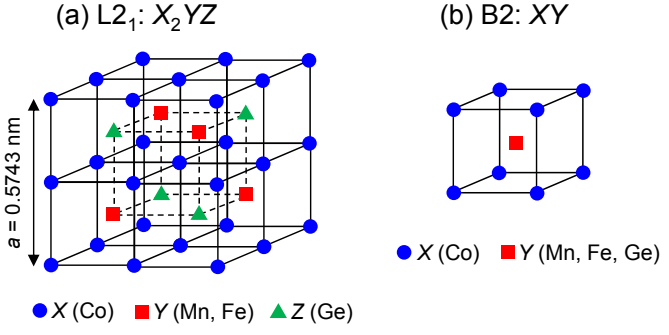

Fig. 1. Unit cells of (a) $L2_1$ structure and (b) B2 structure.