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# Fabrication of magnetic tunnel junctions with a metastable bcc Co<sub>3</sub>Mn disordered alloy as a bottom electrode

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We fabricated MgO barrier magnetic tunnel junctions (MTJs) with a  $Co_3Mn$  alloy bottom and FeCoB top electrodes. The (001)-oriented epitaxial films of the metastable bcc  $Co_3Mn$  disordered alloys obtained showed saturation magnetization of approximately 1640 emu/cm<sup>3</sup>. The transmission electron microscopy showed that the MgO barrier was epitaxially grown on the  $Co_3Mn$  electrode. Tunnel magnetoresistance of approximately 150% was observed at room temperature after the annealing of MTJs at 350°C, indicating that bcc  $Co_3Mn$  alloys have relatively high spin polarization.

The magnetic tunnel junction (MTJ) is a key device for spintronics,  $^{1-3)}$  which has 18 been utilized in various magnetic sensors including the read head of a hard disk drive, 19 magnetoresistive random access memory, and neuromorphic applications.<sup>4–6)</sup> One of the 20 issues is to enhance the tunnel magnetoresistance (TMR) effect, i.e., junction resistance 21 change depending on the parallel and antiparallel states of two magnetizations for the 22 junctions. Currently, the MgO barrier and FeCoB alloy electrodes are used as the stan-23 dard MTJ barrier and magnetic materials,  $^{7-11)}$  which exhibited the record 604% in the 24 TMR ratio at room temperature (RT).<sup>12)</sup> Such a high TMR ratio is attributed to the 25 orbital symmetry filtering by the MgO barrier and the highly spin polarized  $\Delta_1$  band 26 in FeCo alloys.<sup>13,14</sup> To search for routes to further enhance the TMR ratio, it is curious 27 to investigate various magnetic metals other than FeCo binary systems. 28

<sup>29</sup> Here, we report the TMR effect observed in MTJs utilizing different types of dis-

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Fig. 1. Out-of-plane XRD pattern for Co<sub>3</sub>Mn film deposited on (001) Cr-buffered MgO substrate.

ordered bcc CoMn alloy. A bulk Co-rich CoMn binary disordered alloy has a hcp or 1 fcc phase as thermodynamically stable phase.<sup>15–18)</sup> The saturation magnetizations and 2 Curie temperatures decrease with increasing Mn concentration, and a magnetic long 3 range order is lost around the Mn concentration of 30–35%.<sup>15–18)</sup> In contrast, metastable 4 bcc phase of Co-rich CoMn alloys show the relatively high saturation magnetization at 5 similar Mn composition,<sup>19–21)</sup> and a net magnetic moment per atom is in 2.32–2.53  $\mu_B$ 6 at Mn concentration of 24%,<sup>21)</sup> being close to that of a bcc Fe. This bcc phase is ob-7 tained in thin films grown on (001) GaAs and (001) MgO single crystalline substrates by 8 molecular beam epitaxy (MBE) technique, as reported by a few groups.<sup>19–21)</sup> However, 9 there are no reports on MTJs comprised of bcc CoMn alloy electrodes to date. 10

All samples were deposited on (100) MgO single crystal substrates using a magnetron 11 sputtering technique. The base pressure was  $2 \times 10^{-7}$  Pa. The MTJ staking structure was 12 substrate/ Cr(40)/ Co<sub>3</sub>Mn(10)/ Mg(0.4)/ MgO(2)/ Fe<sub>60</sub>Co<sub>20</sub>B<sub>20</sub>(4.5)/ Ta(3)/ Ru(5) 13 (thickness in nm). All layers were deposited at RT. The composition of  $Co_3Mn$  film 14 is  $Co_{74}Mn_{26}$  (at.%) determined using inductively-coupled plasma mass spectrometer. 15 We also prepared samples of substrate/ Cr(40)/ Co<sub>3</sub>Mn(10)/ Mg(0.4)/ MgO(2)/ Ta(2) 16 for structural and magnetization measurements. The crystal structures of the samples 17 were determined using an x-ray diffractometer (XRD) by Cu  $K_{\alpha}$  radiation. Nanostruc-18 tural analysis of samples was conducted by transmission electron microscopy (TEM). 19 Magnetization measurements were performed using a vibrating sample magnetometer. 20



**Fig. 2.** In-plane magnetization hysteresis loop for the Co<sub>3</sub>Mn film deposited on (001) Cr buffered MgO substrate.

The microfabrication of the MTJs were performed using a standard ultraviolet photolithography and Ar ion milling. The thirty six junctions with rectangular shapes were obtained on the substrate with the junction areas of  $60 \times 15$ ,  $40 \times 10$ ,  $20 \times 5$ ,  $40 \times 2$ ,  $15 \times 3$ , and  $20 \times 2 \ \mu m^2$ . The MTJs were annealed with a vacuum furnace at the temperature range 250–400°C. Magnetoresistance (MR) for the MTJs was measured by a four-probe method using a prober system with a maximum applied field of approximately 1 kOe. All the measurements were performed at RT.

<sup>8</sup> Out-of-plane XRD pattern of the Co<sub>3</sub>Mn film is shown in Fig. 1. The 002 peaks from <sup>9</sup> the Cr buffer layer and bcc Co<sub>3</sub>Mn were observed, but no other peaks, in particular <sup>10</sup> those from fcc Co-Mn, were detected. The out-of-plane lattice parameter for the Co<sub>3</sub>Mn <sup>11</sup> film was evaluated as approximately 0.286 nm, which is close to the lattice constant <sup>12</sup> for the bcc Co<sub>3</sub>Mn of 0.285 nm.<sup>19</sup> Thus, it is considered that the (001)-oriented bcc <sup>13</sup> Co<sub>3</sub>Mn films were obtained on (001) Cr-buffered MgO substrates.

The in-plane magnetization curve is shown in Fig. 2. The saturation magnetization  $M_{\rm s}$  is approximately 1640 emu/cm<sup>3</sup>. This value is comparable to that of Co or Fe and is also similar to the magnetic moment value evaluated by x-ray magnetic circular dichroism for bcc Co<sub>76</sub>Mn<sub>24</sub> alloy films,<sup>21)</sup> rather than that of fcc CoMn alloys with the similar Mn concentration.<sup>16)</sup>

<sup>19</sup> The MR curves measured at RT for the  $40 \times 2 \ \mu m^2$ . MTJ annealed at 350°C is shown <sup>20</sup> in Fig. 3(a). The resistance changes depending on the magnetization configuration are



**Fig. 3.** (a) The typical MR curve for MTJs with  $Co_3Mn$  film as the bottom electrode. (b) The TMR ratio as a function of the annealing temperature of the MTJ with  $Co_3Mn$  film as the bottom electrode.

observed. Note that the MTJ is a pseudo-spin valve type, which means that both
magnetic layers were unpinned by the exchange bias<sup>12)</sup> and the antiparallel state would
not be well defined in this study. Figure 3(b) shows the TMR ratio for this junction
as a function of the annealing temperature of the MTJ. The maximum TMR ratio was
observed as 155% at the annealing temperature of 350°C in Fig. 3(b) and was 158%
for the different MTJ on the same substrate. This value is smaller than the TMR ratio
of ~200% observed at RT in Fe/MgO/Fe fully-epitaxial MTJs fabricated by the MBE

 $_1$  technique.<sup>22)</sup>

Figure 4 shows the cross sectional TEM image for the MTJ sample annealed at 3 350°C. The MgO barrier is epitaxially grown on the bcc (001) Co<sub>3</sub>Mn electrode. More-4 over, the coherency of the lattices of Co<sub>3</sub>Mn, MgO, and almost crystallized FeCoB at 5 the bottom and top interfaces are visible. These observations mean that the coherent 6 tunneling is expected if the bcc Co<sub>3</sub>Mn has the  $\Delta_1$  band at the Fermi level.

To gain insight into the spin polarization of the bcc Co<sub>3</sub>Mn studied here, Julliere's
model was used for approximate estimation, which can be expressed as<sup>1)</sup>

TMR ratio (%) = 
$$\frac{2P_1P_2}{1 - P_1P_2} \times 100,$$
 (1)

where  $P_1$  and  $P_2$  are the tunneling spin polarization for each magnetic electrode. Since 9 this relation is hold only for an incoherent tunneling, the evaluate spin polarization 10 should be regarded as an effective value in case of the coherent tunneling. To account 11 for the TMR ratio observed in this study using this relation, the tunneling spin polar-12 ization for bcc  $Co_3Mn$  with MgO barrier should be at least 0.44 at RT if the tunneling 13 spin polarization of FeCoB is 1. This is relatively higher than the spin polarization 14 of 0.33 evaluated at low temperature in  $Co_{73}Mn_{27}$  alloy, which had a low saturation 15 magnetization and was unlikely bcc phase.<sup>23)</sup> A more detailed discussion is beyond the 16 scope of this brief report and will be provided elsewhere. 17

In summary, we fabricated Co<sub>3</sub>Mn/MgO/FeCoB MTJs using the sputtering technique. The (001)-oriented metastable bcc Co<sub>3</sub>Mn epitaxial films obtained exhibited saturation magnetization of approximately 1640 emu/cm<sup>3</sup>. The cross-sectional TEM showed that the MgO barrier was epitaxially grown on the Co<sub>3</sub>Mn electrode. We observed the TMR ratio of 158% at RT for MTJs annealed at 350°C, indicating that metastable bcc Co<sub>3</sub>Mn alloys have relatively high spin polarization.

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Fig. 4. The cross-sectional TEM image for the MTJ sample.

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