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# Effect of a MgO interlayer on the structural and magnetic properties of $\text{Co}_2\text{Cr}_{0.6}\text{Fe}_{0.4}\text{Al}$ thin films epitaxially grown on GaAs substrates

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The effect of a MgO interlayer on the structural and magnetic properties of  $\text{Co}_2\text{Cr}_{0.6}\text{Fe}_{0.4}\text{Al}$  (CCFA) thin films epitaxially grown on GaAs substrates by sputtering was investigated. When the MgO interlayer thickness was 1.0 nm or less, the CCFA films were grown with a cube-on-cube relation to GaAs and no significant decrease in either the x-ray diffraction intensity or saturation magnetization was observed. In contrast, when the MgO thickness was 1.5 nm or more, the CCFA film was rotated by  $45^\circ$  in the (001) plane with respect to GaAs, and both the x-ray diffraction intensity and saturation magnetization decreased. All samples showed strong magnetic anisotropy, in which a uniaxial anisotropy with an easy axis of  $[110]_{\text{GaAs}}$  or  $[1-10]_{\text{GaAs}}$  dominated with a slight cubic anisotropy having an easy axis of  $\langle 110 \rangle_{\text{CCFA}}$  superimposed. © 2007 American Institute of Physics. [DOI: 10.1063/1.2712164]

## I. INTRODUCTION

Co-based full-Heusler alloy thin films are promising ferromagnetic electrode materials for spintronic devices because of their intrinsically high spin polarization at room temperature (RT). Relatively high tunnel magnetoresistance (TMR) ratios were recently observed in magnetic tunnel junctions (MTJs) using Co-based full-Heusler alloys such as  $\text{Co}_2(\text{Cr},\text{Fe})\text{Al}$  (CCFA) (Ref. 1) and  $\text{Co}_2\text{MnSi}$  (CMS).<sup>2,3</sup> In experiments on spin injection from Heusler materials into semiconductors, however, relatively low spin injection efficiency was achieved.<sup>4,5</sup> One reason for the low efficiency could be the formation of a magnetically dead layer due to an interface reaction between Heusler materials and semiconductors.<sup>6</sup> Picozzi *et al.* theoretically predicted from first-principles calculations for  $\text{Co}_2\text{MnGe}$  (CMG)/GaAs and CMG/Ge that the half-metallicity was locally lost at the Heusler material/semiconductor interface.<sup>7</sup> Nagao *et al.* also showed theoretically that the half-metallicity was lost for a (100) interface, but preserved for a (110) interface.<sup>8</sup> Insertion of a thin MgO layer between the Heusler material and the semiconductor is expected to be a useful approach for achieving high spin injection efficiency. In fact, we have successfully demonstrated relatively high TMR ratios in MTJs using a combination of either CCFA,<sup>1,9-11</sup> CMG,<sup>9,12</sup> or CMS,<sup>3</sup> and a MgO tunnel barrier [TMR ratios of about 109% for CCFA-MTJ (Ref. 1) and 90% for CMS-MTJ (Ref. 3) at RT], indicating that the spin polarization at the Heusler material/MgO interface was kept high. In addition, Wang *et al.* demonstrated a high spin injection efficiency of 55% at 100 K in a CoFe/MgO/GaAs structure.<sup>13</sup> Although there have been several reports on the characterization of Heusler materials directly grown on GaAs,<sup>6,14,15</sup> no study on Heusler materials grown on MgO/GaAs has been reported. In this paper, we discuss the effect of a MgO interlayer on the struc-

tural and magnetic properties of CCFA thin films epitaxially grown on GaAs substrates by magnetron sputtering.

## II. EXPERIMENTAL PROCEDURES

Layer structures consisting of 400-nm-thick undoped GaAs and 100-nm-thick *n*-GaAs ( $\text{Si}=3 \times 10^{18} \text{ cm}^{-3}$ ) were grown by molecular beam epitaxy (MBE) at  $580^\circ\text{C}$  on GaAs(001) substrates. Each sample was then capped with an arsenic protective layer and transported in air to an ultrahigh vacuum chamber equipped with magnetron sputtering cathodes and an electron beam (EB) evaporator. Prior to the growth, the arsenic cap was removed by heating the sample to  $400^\circ\text{C}$ . The MgO layer was then grown by EB evaporation, with the thickness ranging from 0 to 2.0 nm in steps of 0.5 nm at  $400^\circ\text{C}$ . Lastly, a 50-nm-thick CCFA film was grown by rf-magnetron sputtering at  $400^\circ\text{C}$ . The crystalline structures of the fabricated CCFA thin films were characterized using x-ray  $\theta$ - $2\theta$  scans and x-ray pole figure measurements (Bruker AXS D8 DISCOVER Hybrid). Magnetic properties were measured using a superconducting quantum interference device (SQUID) magnetometer (Quantum Design MPMS).

## III. RESULTS AND DISCUSSION

### A. Structural properties

Figure 1 shows the pole figures of 022 diffraction for the CCFA thin films with MgO thickness ( $t_{\text{MgO}}$ ) of (a) 0, (b) 1.0, and (c) 1.5 nm. Here, we set the GaAs[110] direction to the origin of  $\phi$ . The CCFA 022 diffraction peaks showed a four-fold symmetry with respect to the sample rotation angle  $\phi$  at a tilt angle  $\chi$  of  $45^\circ$  in both samples; this was the direct evidence of epitaxial growth of single-crystalline CCFA films deposited on GaAs substrates with a MgO interlayer. The crystallographic relationship with respect to the GaAs was CCFA(001)[110]||GaAs(001)[110], i.e., a cube-on-cube relation for  $t_{\text{MgO}} \leq 1.0 \text{ nm}$  [Figs. 1(a) and 1(b)]. When  $t_{\text{MgO}}$

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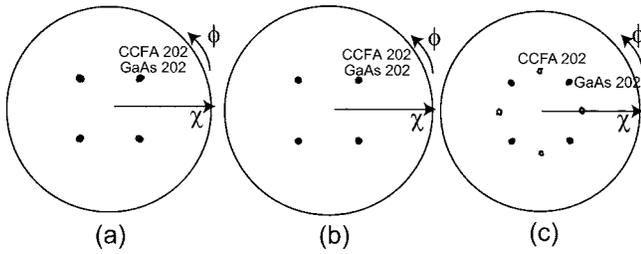


FIG. 1. Pole figures of CCFA 022 diffraction in the samples with (a)  $t_{\text{MgO}} = 0$  nm, (b)  $t_{\text{MgO}} = 1.0$  nm, and (c)  $t_{\text{MgO}} = 1.5$  nm. In these figures, GaAs 022 peaks are also seen due to a small lattice mismatch.

$\geq 1.5$  nm, however, the poles of CCFA 022 diffraction shifted by  $45^\circ$  with respect to those of the GaAs [Fig. 1(c)], indicating that the crystallographic relation was CCFA(001)[110]||GaAs(001)[100]. From this result, we deduced that the MgO was grown on GaAs with a cube-on-cube structure and was relaxed when  $t_{\text{MgO}} \geq 1.5$  nm.

Figure 2 shows the x-ray diffraction intensity of the CCFA 022 peak as a function of  $t_{\text{MgO}}$ . The intensity was drastically decreased when  $t_{\text{MgO}} \geq 1.5$  nm. The B2 structure was dominant when  $t_{\text{MgO}} \leq 1.0$  nm, since we observed 002 and 222 diffractions but no 111 diffraction. When  $t_{\text{MgO}} \geq 1.5$  nm, on the other hand, we observed only 022 diffraction, indicating that the A2 structure, which is more disordered than the B2 structure, was dominant.

## B. Magnetic properties

Figure 3 shows the saturation magnetization ( $M_S$ ) at RT as a function of  $t_{\text{MgO}}$ . The  $M_S$  of the CCFA without a MgO layer was  $2.6\mu_B/\text{f.u.}$ , a value comparable to that reported for MBE-grown CCFA films ( $2.3\mu_B/\text{f.u.}$  at RT).<sup>14</sup> As  $t_{\text{MgO}}$  increased,  $M_S$  gradually decreased. Miura *et al.* recently calculated atomic disorder effects on the half-metallicity of CCFA and showed that the A2-type disorder induced a significant reduction of the magnetic moment as well as spin polarization.<sup>16</sup> The reduction of  $M_S$  shown in Fig. 4 was therefore probably due to the increased atomic disorder, because the A2 structure was dominant when  $t_{\text{MgO}} \geq 1.5$  nm.

Figure 4 shows the magnetic hysteresis curves for samples with  $t_{\text{MgO}}$  of (a) 0 nm and (b) 1.5 nm. The magnetic field was applied along the GaAs [110], [1-10], and [100] axes. All samples showed strong magnetic anisotropy. This

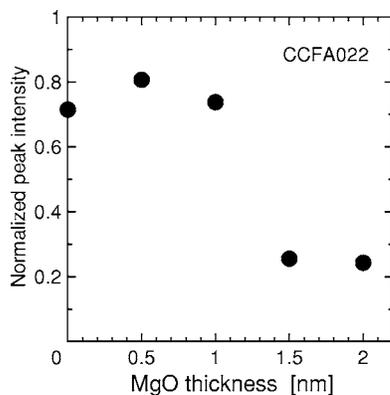


FIG. 2.  $t_{\text{MgO}}$  dependence of CCFA 022 diffraction peak intensity.

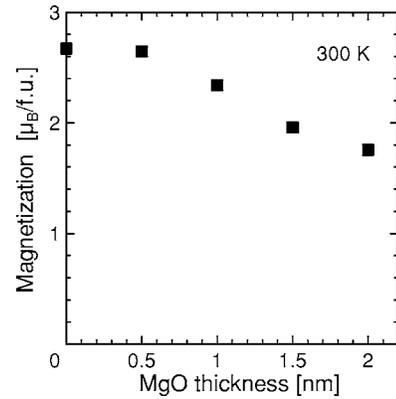


FIG. 3. Dependence of saturation magnetization of CCFA films on  $t_{\text{MgO}}$  at RT.

contrasted with the result for the reference CCFA films grown on a MgO substrate, in which almost no uniaxial magnetic anisotropy was observed.

The anisotropy shown in Fig. 4(a) was similar to that observed in MBE-grown CCFA (Ref. 14) or other Heusler materials on GaAs.<sup>15,17</sup> This can be explained with the model in which (i) uniaxial anisotropy imposed by the GaAs symmetry with an easy axis of either the  $[110]_{\text{GaAs}}$  or  $[1-10]_{\text{GaAs}}$  direction and (ii) cubic anisotropy imposed by the CCFA with easy axes of  $\langle 110 \rangle_{\text{CCFA}}$  are superimposed.<sup>17,18</sup> In our case, the uniaxial anisotropy with its easy axis in the  $[110]_{\text{GaAs}}$  direction was dominant compared to the cubic anisotropy, as we will later show. The magnetic hard axis direction, therefore, was along  $[1-10]_{\text{GaAs}}$ .

The CCFA with a 1.5-nm-thick MgO layer also showed strong magnetic anisotropy [Fig. 4(b)]. The magnetic hysteresis curves differed in shape from those shown in Fig. 4(a) because of the difference in the crystallographic relations. To confirm this and to evaluate the anisotropy constants in both samples, we simulated the hysteresis curves taking into consideration both the in-plane cubic and the uniaxial anisotropy energies. The free energy density with combined cubic anisotropy and uniaxial anisotropy can be modeled as

$$E = \frac{K_1}{4} \sin^2 2(\varphi - \alpha) + K_u \sin^2(\varphi - \beta) - MH \cos(\varphi - \theta), \quad (1)$$

where  $K_1$  and  $K_u$  are cubic and uniaxial anisotropy constants,  $\alpha$  and  $\beta$  are the easy axis directions for cubic and uniaxial

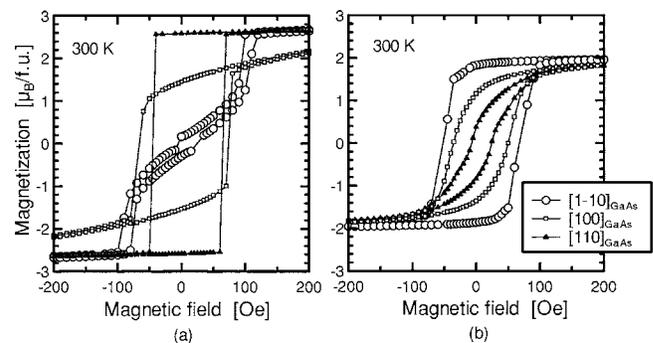


FIG. 4. Magnetic hysteresis curves for the CCFA films with (a)  $t_{\text{MgO}} = 0$  nm and (b)  $t_{\text{MgO}} = 1.5$  nm.

TABLE I. Easy axis directions and anisotropy constants for CCFA on GaAs with and without MgO.

	$t_{\text{MgO}}=0$ nm	$t_{\text{MgO}}=1.5$ nm
$\alpha$ (deg)	$0^\circ$ ( $\langle 110 \rangle_{\text{GaAs}}$ )	$45^\circ$ ( $\langle 100 \rangle_{\text{GaAs}}$ )
$\beta$ (deg)	$0^\circ$ ( $[110]_{\text{GaAs}}$ )	$90^\circ$ ( $[1-10]_{\text{GaAs}}$ )
$K_1$ (ergs/cm <sup>3</sup> )	$2.2 \times 10^4$	$1.1 \times 10^4$
$K_u$ (ergs/cm <sup>3</sup> )	$3.7 \times 10^4$	$1.8 \times 10^4$

anisotropies with respect to the  $[110]_{\text{GaAs}}$  direction,  $M$  is the magnetization,  $H$  is the magnetic field, and  $\varphi$  and  $\theta$  are the directions of  $M$  and  $H$ .

Table I shows the deduced easy axis directions and anisotropy constants for both samples. The value of  $\alpha$ , the easy axis direction for the cubic anisotropy, in the CCFA with  $t_{\text{MgO}}$  of 1.5 nm differed by  $45^\circ$  from that in the CCFA without MgO, while the value of  $\beta$ , the easy axis direction for the uniaxial anisotropy, differed by  $90^\circ$ . Since the direction of cubic anisotropy was  $\langle 110 \rangle_{\text{CCFA}}$  for both samples, we attribute its origin to the crystal symmetry of the CCFA. The origin of the uniaxial anisotropy, on the other hand, remains an open question. As for the sample without a MgO interlayer, several mechanisms relating to the antisymmetry of the GaAs substrate, such as a surface reconstruction of GaAs, formation of an interface alloy, or anisotropic interfacial bonds, may cause the uniaxial anisotropy, as was discussed in (Ref. 6). Interestingly, we found in this study that the uniaxial anisotropy of the CCFA existed even in the presence of the thin MgO interlayer and that the easy axis direction changed by  $90^\circ$  depending on  $t_{\text{MgO}}$ . Its origin is more complicated. Since no uniaxial anisotropy was observed for the CCFA grown on a MgO substrate, the GaAs substrate plays an important role in inducing the anisotropy even in the presence of the MgO interlayer. A large lattice mismatch of approximately 25% exists between GaAs and MgO and the easy axis direction was closely related to whether the MgO layer was strained or relaxed. Thus, a strain or a strain-induced disorder is one possible mechanism accounting for the uniaxial anisotropy of the sample with the MgO interlayer. To fully understand its origin, though, further systematic studies are necessary.

The magnetic anisotropy constants shown in Table I were deduced by fitting the experimental data for the hard axis directions ( $[1-10]_{\text{GaAs}}$  for the CCFA without MgO and  $[110]_{\text{GaAs}}$  for the CCFA with 1.5-nm-thick MgO) using the Stoner-Wohlfarth formation for coherent rotation reversal, since the coherent rotation reversal is dominant when the magnetic field is applied along the hard axis direction, while domain-wall motion inevitably occurs for other directions. Figure 5 shows the simulated results alongside the experimental data. Note that since we could not apply the field along the exact hard axis in the experiment, we assumed in the simulation that the field was applied off the hard axis by  $3^\circ$ – $5^\circ$ . The close agreement between the experiment and simulation results indicates that the assumed magnetoanisotropy model is reasonable. The uniaxial anisotropy constants were approximately 1.6–1.7 times as large as the cubic an-

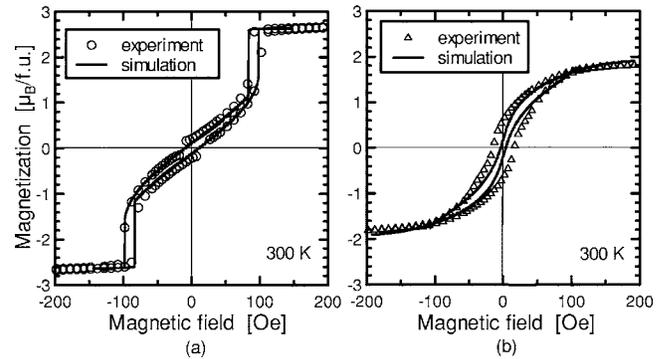


FIG. 5. Magnetic hysteresis curves along the hard axis direction for the CCFA films with (a)  $t_{\text{MgO}}=0$  nm and (b)  $t_{\text{MgO}}=1.5$  nm. Circles indicate the experimental data and solid lines indicate simulation.

isotropy constants for both samples, indicating that the uniaxial anisotropy was dominant compared to the cubic anisotropy.

#### IV. SUMMARY

We have successfully grown epitaxial CCFA films on GaAs substrates with a MgO interlayer. When the MgO interlayer thickness was 1.0 nm or less, the CCFA films were grown with a cube-on-cube relation to GaAs, and no significant decrease of either the x-ray diffraction intensity or the saturation magnetization was observed. This appears to be a useful approach to realize high spin injection efficiency. When the MgO thickness was 1.5 nm or more, though, the CCFA films were rotated by  $45^\circ$  in the (001) plane with respect to GaAs probably due to the lattice relaxation of MgO layer, and both the x-ray diffraction intensity and the saturation magnetization decreased. All samples showed strong magnetic anisotropy, and the uniaxial magnetic anisotropy that originated from the antisymmetry of the GaAs was dominant even in the presence of the thin MgO interlayer. We found that the strong uniaxial magnetic anisotropy existed even in the presence of the MgO interlayer and that the easy axis direction changed depending on the MgO thickness.

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