## Magnetic anisotropy switching in heavily-Fe-doped high-Curie-temperature ferromagnetic semiconductor (Ga0.7,Fe0.3)Sb with a critical thickness

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Magnetic anisotropy (MA) plays an important role in determining the magnetization direction of ferromagnetic materials in spintronics devices. For Mn-doped III-V-based ferromagnetic semiconductor (FMS) (Ga,Mn)As, the thickness dependence of the uniaxial anisotropy fields has been well investigated [1]. However, due to the low Curie temperature ( $T_C < 200$ K), (Ga,Mn)As is not suitable for practical device applications. To overcome this problem, recently we have successfully grown Fe-doped FMS (Ga,Fe)Sb, which shows a high  $T_C$  (> 300 K) when the Fe concentration is higher than 23% [2]. Therefore, this new material is a good candidate for device applications operating at room temperature.

Recently, we investigated the MA of  $(Ga_{1-x}, Fe_x)Sb$  (x = 0.2, thickness d = 15 nm) and found that in-plane magnetic anisotropy (IMA) is dominant due to the large shape anisotropy [3]. In this work, to control the MA, we have investigated the thickness dependence of the MA of  $(Ga_{1-x}, Fe_x)Sb$  (x = 0.3, d = 20, 30, 40, and 55 nm) grown by low-temperature molecular-beam epitaxy (LT-MBE) on AISb using semi-insulating GaAs(001) substrates. Figure 1(a) show the schematic sample structure of  $Ga_{0.3}Fe_{0.7}Sb$  (d= 40nm). All the samples showed  $T_{\rm C} > 320$  K. We estimated the MA constants by measuring ferromagnetic resonance (FMR) and magnetization using a superconducting quantum interference device (SQUID) at room temperature. Fig. 1(b) shows the FMR spectra measured at 300 K for the samples with d = 20 nm and 55 nm, respectively, when the magnetic field H is applied along the in-plane [110] and the perpendicular [001] directions. Figure 1(c) shows the resonance field as a function of the out-of-plane direction of H in the (Ga,Fe)Sb films with various d. The same FMR measurements were performed applying H along various directions in the film plane. By fitting to the H-angle dependence of the resonance field combining with the value of  $\mu_0 M_{\rm S}$  measured by SQUID, we estimated the perpendicular uniaxial anisotropy constant  $(K_{2\perp})$ , the shape anisotropy constant  $(K_{sh})$ , the perpendicular cubic anisotropy constant  $(K_{4\perp})$ , in-plane uniaxial anisotropy constant  $(K_{2//})$ , and the effective anisotropy constant  $(K_{eff} = K_{2\perp} + K_{sh} + K_{4\perp} + K_{2//})$  of all the (Ga,Fe)Sb thin films. In this work, the negative sign of  $K_{\rm eff}$  represents the IMA, while the positive sign corresponds to the perpendicular magnetic anisotropy (PMA). To analyze the thickness dependence of  $K_{\rm eff}$ , we separated the bulk and interface contributions in  $K_{\text{eff}}$  by using the following linear equation:  $K_{\text{eff}}d = K_{\text{V}}d$  $+K_{\rm S}$  where,  $K_{\rm V}$  is the bulk contribution and  $K_{\rm S}$  is the interface contribution in  $K_{\rm eff}$ . Figure 1(d) summarizes the obtained  $K_{eff}d$  as a function of d. On fitting by linear equation, we obtain  $K_S$  in negative sign which causes IMA whereas  $K_V$  is positive sign which causes PMA in our (Ga,Fe)Sb films.

In Fig. 1(d), when d is changed from 20 nm to 55 nm, the sign change of  $K_{\text{eff}d}$  from negative (IMA) to positive (PMA) occurs. When d is small (d = 20, 30, and 40 nm), the IMA originates from the film interface due to the tensile strain that is dominant when the film is thin. This result is consistent with our previous report where the films have tensile strain when grown over AlSb buffer layer [3]. In contrast, with increasing d (d = 55 nm), PMA becomes dominant from the bulk contribution, which may be due to the columnar-like growth occurring when d is larger than 30 nm. Interestingly, we did not observe cubic anisotropy in (Ga,Fe)Sb and thus only two-fold symmetry along the [ $\overline{110}$ ] axis exists. These results are different from those of (Ga,Mn)As, which has four-fold symmetry due to its dominant cubic anisotropy along the <100> axes. The observation of FMR at room temperature and the control of magnetic anisotropy are important steps toward device applications of (Ga,Fe)Sb.

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Fig.1 (a) Schematic sample structure of  $(Ga_{0.7}Fe_{0.3}Sb)$ . (b) FMR spectra observed for  $Ga_{0.7}Fe_{0.3}Sb$  with d = 20 nm and 55 nm at 300 K when the magnetic field *H* was applied along the in-plane [110] (red circles) and the perpendicular [001] axes (black squares). (c) FMR field as a function of the *H* direction  $\theta_{\rm H}$  (where the inset shows definitions of  $\theta_{\rm H}$  and  $\theta_{\rm M}$ , which are the applied magnetic field angle and magnetization angle, respectively. (d)  $K_{\rm eff} (=K_{2\perp}+K_{\rm sh}+K_{4\perp}+K_{2//})$  times *d* as a function of *d* of (Ga<sub>0.7</sub>,Fe<sub>0.3</sub>)Sb.

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