Single crystal Fe films grown on Ge (001) substrates by magnetron sputtering

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Single crystal Fe films were grown on Ge (001) substrates by using dc magnetron sputtering. It was found that the microstructures and magnetic properties of Fe films on Ge substrates were strongly dependent upon the substrate temperature during the deposition process. There existed a narrow substrate temperature window of 125±25 °C for achieving single crystal Fe film on Ge. Lower substrate temperature led to polycrystalline Fe films due to limited mobility of Fe atoms, while higher substrate temperatures resulted in amorphous Fe–Ge alloy due to severe interdiffusion.
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Successful integration of magnetic films onto semiconductor substrates is crucial for many applications, such as integrated magnetic microwave devices[1,2] and spintronics applications.[3] Single crystal Fe thin films on GaAs substrates have been grown by molecular beam epitaxy (MBE) and ion-beam sputtering.[1,4–7] The lattice constant of GaAs (a_0=5.653 Å) is very close to twice of that of bcc Fe (a_0=2.867 Å) with a lattice mismatch of 1.4%, which leads to epitaxial growth of Fe thin films. Single crystal Fe thin films on other semiconductor substrates, such as ZnSe (Ref. 8) and InAs,[9] were also reported.

Another interesting magnetic/semiconductor system is the Fe/Ge system. The Ge lattice (a_0=5.658 Å) and Fe lattice have a small lattice mismatch of 1.3%, which makes Ge substrates suitable for growing single crystal Fe films. However, growing single-crystal Fe films on Ge has been challenging due to the severe intermixing/diffusion between Fe and Ge at high temperatures, which causes thick magnetiv dead layers up to 100 Å[10,11] Most previous efforts on growing single crystal Fe film on Ge substrates were carried out by MBE.[11,12]

In this letter, single crystal Fe films were grown on Ge (001) substrates by dc magnetron sputtering. Substrate temperature dependence of the microstructures and magnetic properties of the Fe films on Ge substrates were systematically studied.

Intrinsic Ge (001) substrates with a resistivity of 35 Ω cm were used. A three-step cleaning process was employed for Ge substrates. The first step was an ultrasonic cleaning in acetone and alcohol, both for 10 min; the second step was a 5 nm plasma etching; and the last step was a high temperature (500 °C) annealing for removing oxides and improving the surface smoothness. Fe films were deposited with a constant thickness of ~70 nm on Ge substrates by dc magnetron sputtering at a target power density of ~5 W/cm^2 in a 3 mTorr Ar atmosphere. Substrate temperature was varied between room temperature and 300 °C with a base pressure of 1×10^{-8} Torr.

Crystal structures of Fe films were characterized by X-ray diffraction (XRD) with a Cu $K\alpha$ source (λ=1.541 Å), transmission electron microscope (TEM), and a general area detector diffraction system (GADDS). Atomic force microscope (AFM) was used to examine the morphologies of Fe films. Magnetic properties of the Fe films were characterized by vibrating sample magnetometer (VSM) and field-sweep ferromagnetic resonance (FMR) spectrometer at *X* band (~9.5 GHz).

Figure 1 shows the XRD patterns of Fe films grown on Ge substrates at different temperatures. The Ge (004) diffraction peak was used for calibrating the diffraction peak intensities and positions of all samples. The Fe film deposited at a substrate temperature of 75 °C shows only one broad bcc Fe

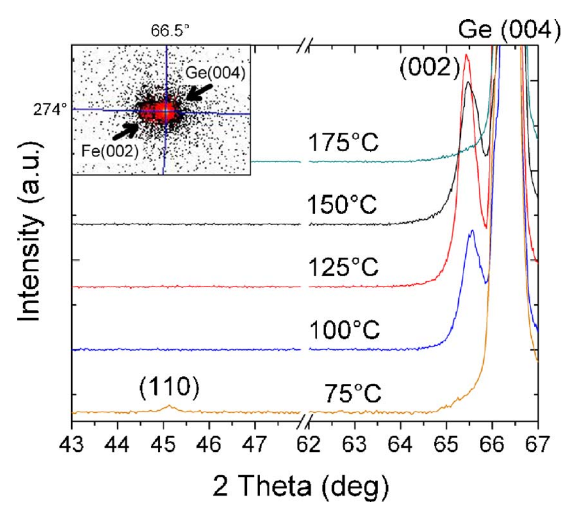

FIG. 1. (Color online) XRD patterns of Fe films grown at (a) 75 °C, (b) 100 °C, (c) 125 °C, (d) 150 °C, and (e) 175 °C. The top-left insert is the GADDS pattern of (c).

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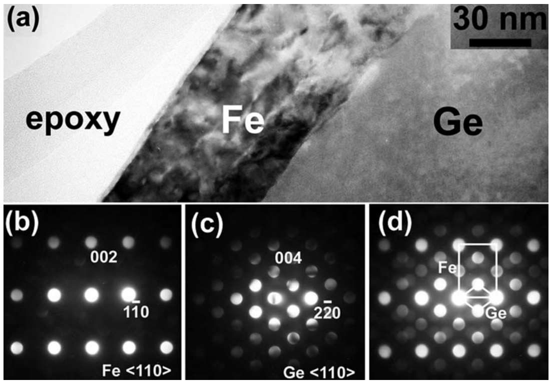

FIG. 2. (a) Cross-section TEM image of the Fe film grown at 125 °C and its converged beam diffraction patterns at (b) Fe film, (c) Ge substrate, and (d) Fe–Ge interface.

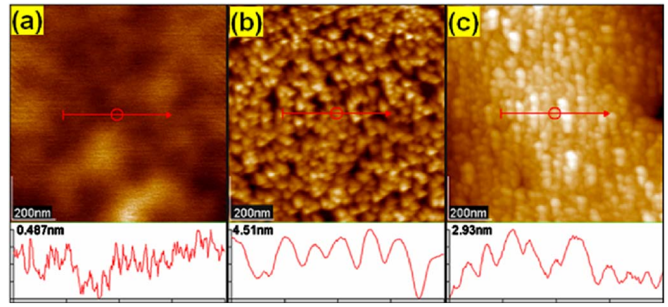

FIG. 3. (Color online) AFM images of Fe films deposited at (a) 125 °C, (b) 75 °C, and (c) 175 °C.

(110) peak with a low intensity, indicating polycrystalline Fe film growth on Ge substrates. The bcc Fe (002) peak appears for the Fe samples grown at 100, 125, and 150 °C, which indicates that there are Fe (001) planes parallel to the Ge (001) planes, implying the possibility of epitaxial growth of Fe films on Ge. At 125 °C, the Fe (002) peak intensity is more than twice of those grown at 100 and 150 °C. When the substrate temperature reaches 175–300 °C, there was no Fe diffraction peak, while a thick layer (up to 100 nm) of amorphous Fe–Ge alloy film with a nearly constant Ge content of up to ~30 at. % was observed on the film top by x-ray photoelectron spectroscopy depth profile analyses.[13] Obviously, heavy intermixing or interdiffusion between the Fe film and Ge substrates occurred at high substrate temperatures, as reported in literature.[11]

The X-ray diffraction pattern from GADDS for the Fe film deposited at 125 °C is shown in the top-left insert of Fig. 1, too. Only two strong distinct diffraction peaks were observed, which were indexed to be Fe (002) and Ge (004) peaks as indicated in the chart. The φ angles of both Fe (002) peak and Ge (004) are very well aligned to each other, confirming that the Fe film on Ge was epitaxial with an orientation relationship of Fe {002}‖Ge {004}.

Cross-section TEM imaging was done on the Fe film grown at 125 °C. A well-defined Fe/Ge interface was observed, as shown in Fig. 2. The converged beam diffraction patterns on Fe film side, Ge side, and Fe/Ge interface clearly show the single crystal nature and the orientation relationship between the Fe film and Ge substrate, which is ⟨110⟩Fe‖⟨110⟩Ge and {002}Fe‖{004}Ge, as indicated in Figs. 2(b)–2(d). The TEM diffraction patterns are obviously consistent with the GADDS as well as XRD results, showing the epitaxial nature of the Fe films on Ge substrates.

Figure 3(a) shows the typical AFM image of the Fe film grown at 125 °C within a 1×1 μm^2 region, which exhibits a smooth surface with low roughness (R_a) of 0.21 nm. The Fe film grown at 75 °C, however, shows a much rougher surface with a roughness of 0.78 nm, as shown in Fig. 3(b). The surface is no longer continuous with nearly columnar structures. When the substrate temperature was increased to 175 °C, the Fe film surface got rougher again with a roughness of 1.2 nm as shown in Fig. 3(c). The low roughness for the Fe film grown at 125 °C correlates well with the epitaxial nature of the Fe film as shown by the XRD, GADDS, and TEM.

Figure 4(a) shows hysteresis loops of the Fe film grown at 125 °C when the external fields were applied along the ⟨110⟩ and ⟨100⟩ directions of Ge substrate, i.e., along the Fe ⟨110⟩ and Fe ⟨100⟩ orientations. The hysteresis loops show a remanence of 74% (see dashed line) along Fe ⟨110⟩, and 97% along Fe ⟨100⟩ with a relatively low coercive field of 14 Oe. The same Fe film deposited at 125 °C was an-

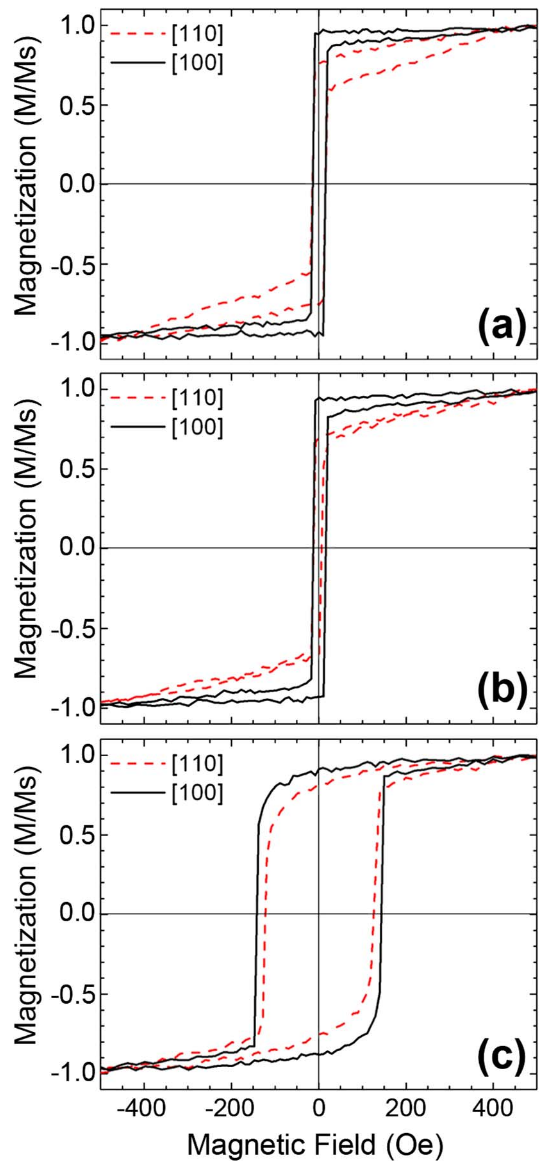

FIG. 4. (Color online) Hysteresis loops of Fe films grown at (a) 125 °C, as deposited, (b) 125 °C, after annealing at 125 °C for 6 h, and (c) 75 °C, as deposited.

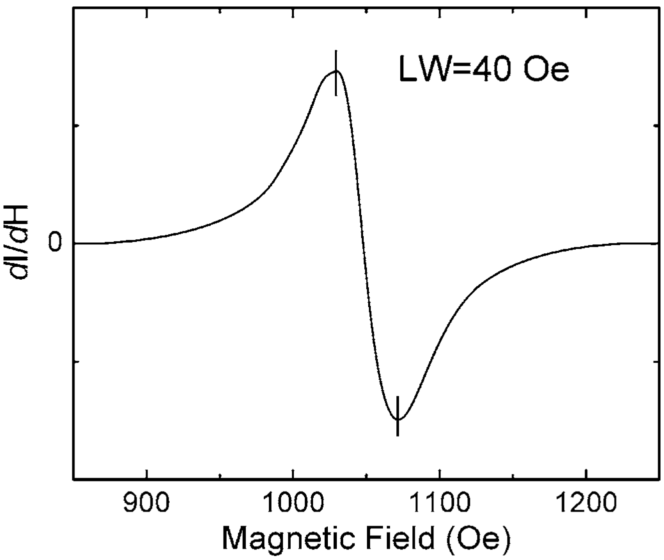

FIG. 5. *X*-band FMR spectrum of the Fe film grown on Ge substrate at 125 °C.

nealed in vacuum at 125 °C for 6 h; the hysteresis loops are improved with reduced coercivity, and closer to those of ideal single crystal Fe, as indicated in Fig. 4(b) with a remanence of 70% along Fe ⟨110⟩, which is closer to the theoretical value of 70.7% for ideal single crystal Fe film. The hysteresis loops shown in Figs. 4(a) and 4(b) are clearly consistent with the orientation relationship observed by TEM. In contrast, the Fe film grown at 75 °C [Fig. 4(c)] shows much higher coercivities of ~140 Oe without obvious in-plane anisotropy, which is consistent with the polycrystalline nature of the film.

FMR measurements on the 70 nm thick Fe films grown at different temperatures were carried out at *X* band (~9.5 GHz) with external fields applied along the Ge ⟨110⟩ orientation, i.e., the magnetic hard axis direction of the Fe film. No FMR peak was observed when the field was applied along the Ge ⟨100⟩ orientation for the same sample due to the large magnetocrystalline anisotropy field along the Fe ⟨100⟩ easy axis. Minimum FMR linewidth of 40 Oe was obtained for the 70 nm thick Fe films deposited at a substrate temperature of 125 °C (see Fig. 5). The reported narrow FMR linewidths at *X* band for single crystal Fe films is ~13 Oe for a 13.6 nm thick Fe on ZnSe (001),[8] and 12–15 Oe for Fe films with a thickness of 8–15 nm on MgO (001) substrates.[14] It is notable that the FMR linewidth of metal magnetic films is a strong function of the film thickness due to different reasons, such as surface anisotropy, defect density, and eddy currents. For example, for high quality single crystal Fe films on MgO substrate, the FMR linewidth at *X* band is ~15 Oe at a thickness of 8 nm, which increases to 45 Oe at an Fe film thickness of 75 nm.[14] The FMR linewidth of 40 Oe for the 70 nm thick Fe film on Ge substrate grown at 125 °C is comparable to those published FMR linewidth values of the Fe films of similar thickness.[14]

It is interesting to note that despite the fact that the Fe film deposited at 125 °C showed improved hysteresis loops after annealing at 125 °C for 6 h, the FMR linewidth of the annealed Fe film was increased from 40 to 55 Oe. Depth profile analysis indicated that severe intermixing between Fe and Ge occurred after a long time annealing at 125 °C.[13,15] This intermixing between Fe and Ge poses a great challenge on achieving high quality single crystal Fe films on Ge substrates.

It is clear that the microstructures and magnetic properties of Fe films on Ge substrates are highly dependent on the substrate temperature during the deposition process. There exists a very narrow substrate temperature window of 125±25 °C for achieving single crystal Fe films on Ge substrates. When the substrate temperature is low (<100 °C), the mobility of Fe adatoms is limited, which leads to polycrystalline Fe films; however, when the temperature is too high (>150 °C), the mobility of Fe adatoms as well as Ge atoms from the substrate are enhanced, leading to an amorphous Fe–Ge alloy film.[13,15] Either too low or too high a mobility of the Fe adatoms should be avoided for growing single crystal Fe films on Ge on Ge substrates.

In conclusion, it was found that single crystal Fe films on semiconductor Ge (001) substrates can be grown within a narrow temperature window [125±25 °C]. This was done with the low-cost dc magnetron sputtering at a relatively low substrate temperature that is compatible with complementary metal-oxide-semiconductor (CMOS)/bipolar CMOS and monolithic microwave integrated circuit technologies. All XRD, TEM, VSM, as well as FMR results confirmed the epitaxial nature of single crystal Fe thin films on Ge substrates.

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