Modification of the Microstructure and Magnetic Properties of Ga-Ion-Irradiated Co/Pd Multilayer Film for Future Planar Media

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Ga ion irradiation was used to modify the microstructures and magnetic properties of Co/Pd multilayers, and also to fabricate patterned media. A decrease in the diffraction peak at low-angle XRD showed that the regular interface between Co and Pd layers was blurred by ion irradiation. The effective perpendicular magnetic anisotropy energy, K_{eff} , decreased from 1.7×10^6 erg/cm³ for as-grown film to 0.4×10^6 erg/cm³ for irradiated film at 2×10^{15} ions/cm². The easy axis of magnetization changed from perpendicular to in-plane at the highest ion dose (5×10^{15} ions/cm²). An MFM image showed that the domain width gradually decreased with increasing ion dose, which corresponds to a decrease in the perpendicular magnetic anisotropy. Sharp boundaries between perpendicular and in-plane regions are also observed in patterned films, which indicates that localized magnetically patterns can be engineered by Ga ion irradiation.

Key words: planar media, Co/Pd multilayer film, ion irradiation, perpendicular magnetic anisotropy, magnetic domain

1. Introduction

In the last several years, the rapidly growing interest in nano-scale magnetic thin film and patterned recording media based on lithographically patterned structures has made them promising base elements for future magnetic recording media¹⁻⁵, since the increasing density of conventional recording media will be limited by superparamagnetic behavior. Arrays for patterning the magnetic structure may be fabricated by lithography techniques such as etching, lift-off, and electroplating⁶⁻⁸. However, to define the patterning arrays of the data bits, these techniques change the surface roughness of the magnetic media, and they have other drawbacks such as poor planarity. This would be a problem in hard disk recording technology, where the head flies at very high speed only 10 nm above the surface of the recording medium.

It has recently been reported that magnetic patterning of continuous magnetic film can be achieved by ion beam irradiation without significant modification of the surface roughness of the magnetic films⁹. Chappert *et al.* first reported planar-patterned Co/Pt magnetic multilayer films,

where the He ion irradiation technique was used to induce local chemical mixing in a Co/Pt multilayer film⁹. The patterned magnetic structure resulted from the modulated distribution of chemical mixing, without any significant change in the topography. Using a different dose of high-energy ions, the researchers were also able to effectively reduce the coercivity of Co/Pt multilayers. At a sufficiently high He ion dose of 10^{16} ions/cm², the easy axis of Co/Pt multilayers is rotated from perpendicular into the plane of the film.

Co/Pd multilayer films have also been extensively studied, since they exhibit large perpendicular magnetic anisotropy (PMA) and high coercivity¹⁰⁻¹². It has been reported that ion irradiation caused a decrease in the uniaxial magnetic anisotropy of Co/Pd multilayer films¹³. However, no systematic studies have yet been reported of the process for fabricating planar-patterned magnetic film in Co/Pd multilayer film or of the possible correlation between the magnetic properties of irradiated Co/Pd multilayer films and their microstructure, nor has there been any investigation of how isolated pattern magnetic domain structures can appear in planar-patterned media. Since the irradiated area has the same magnetization as the non-irradiated area, the exchange coupling between the bits may become important when ion irradiation is used to separate bits on the media. In this paper, the effects of Ga ion irradiation on the magnetic properties of Co/Pd multilayer are investigated. Ga ion irradiation produced by a focused ion beam (FIB) is an excellent tool for fabricating a patterned magnetic structure without modification of the sample topography. The influence of ion irradiation on the magnetic properties and microstructure of Co/Pd multilayer films and the modification of the magnetic properties of Co/Pd multilayer films by Ga ion irradiation correlated with domain structure are discussed and analyzed in detail. An array of squares of Co/Pd multilayer film with perpendicular magnetic anisotropy was also patterned by FIB, to study the feasibility of using such arrays in planar patterned media.

2. Experimental method

Pd (10 nm)/[Co (0.4 nm)/Pd (1.35 nm)]₂₀/SiN (70 nm) films were prepared on a silicon substrate by RF magnetron sputtering. The background pressure in the sputtering system was below 3.0×10^{-7} Torr. The Ar

pressure during the sputtering of the Co/Pd multilayer films was set at 20 mTorr. The Co/Pd multilayer films were irradiated with a 22 keV Ga^+ focused ion beam (FIB) at various ion irradiation doses from 5×10^{12} to 5×10^{15} Ga ions/cm² in order to study the corresponding changes in the magnetic properties of irradiated magnetic films. The ion irradiation was performed at room temperature with the Ga ion beam normal to the film surface. The *M-H* loops of the sheet films were measured with an alternating gradient magnetometer (AGM). The magnetic anisotropies of films were measured with a torque magnetometer. Irradiated Co/Pd multilayer films with sides 3 mm square and 10 mm square were prepared for AGM, torque magnetometer, and X-ray diffraction (XRD) measurement. The influence of the ion dose on the domain structure was studied by examining magnetic force microscopy (MFM) images. The surface profile was determined by atomic force microscopy (AFM). The $10 \times 10 \mu\text{m}^2$ squares were irradiated for AFM and MFM measurement. The layered and crystalline structures were analyzed by the X-ray diffraction (XRD) technique, using Cu K_α (40 kV, 30 mA) radiation.

Patterned Co/Pd samples were also fabricated by FIB etching at ion irradiation doses of 5×10^{15} ions/cm². In the patterned films, un-irradiated squares with a size of 1 μm were separated by irradiated regions with a size of 0.7 μm to study the located magnetic domain structures of the planar-patterned magnetic media.

3. Results and discussion

3.1 Microstructure and magnetic properties

Fig. 1 shows the etching depth estimated from the AFM image as a function of the Ga ion dose. The etching depth of films irradiated with the maximum ion dose of 1×10^{16} ions/cm² was about 5.4 nm. Since the thickness of the Pd capping layer is 10 nm, only the Pd capping layer is assumed to be etched by the FIB irradiation, and the Co/Pd magnetic multilayer film is not thought to be oxidized or etched.

Fig. 1. Variation of the etching depth estimated by AFM result as a function of the Ga^+ ions dose. The zero ion dose is for as-grown film.

Fig. 2. Low-angle X-ray diffraction profiles of as-grown film (a) and irradiated films with ion doses of 2×10^{13} (b), 1×10^{14} (c), and 5×10^{14} ions/cm² (d).

Fig. 2 shows a low-angle X-ray diffraction profile of as-grown and irradiated Co/Pd multilayer films with various Ga ion doses. The low-angle X-ray diffraction profile is useful for determining the regularity of the periodic structure of Co/Pd multilayers. The first peak indicated in Fig. 2 is attributed to the periodicity of the bilayer in the Co/Pd multilayer film, which corresponds to the first-order diffraction of Co/Pd bilayers with a periodicity of 1.76 nm as compared to the nominal periodicity of 1.75 nm. The different first peak intensities of the four samples mainly reflect the difference in the interlayer structure of the Co/Pd multilayer film. As-grown film exhibited a well-defined periodic structure and regular interface, as shown in the greater first peak intensity. It is known that the regular interface in the multilayers promotes a surface magnetic anisotropy in the easy axis perpendicular to the film plane. The first peak was decreased and broadened by increasing the ion irradiation dose, and disappeared at 5×10^{14} ions/cm². These results suggest that high-energy Ga ion irradiation diminishes the regular interfaces between Co and Pd layers in Co/Pd multilayer films, on account of intermixing of atoms in the layers. The energy transferred to the multilayers via ion irradiation may affect the local atomic displacements and the interdiffusion of Co and Pd atoms.

Fig. 3 shows the *M-H* loops of as-grown and irradiated Co/Pd multilayer films when a magnetic field is applied perpendicular and parallel to the film plane. As-grown Co/Pd multilayer film was confirmed to exhibit a square-shaped *M-H* loop and display perpendicular easy magnetization with a remanence ratio of about 1 and a coercivity value of 1.45 kOe, as shown in Fig. 3(a). The as-grown sample showed an *H_c* value of 1.45 kOe, but it has a rather large uniaxial anisotropy of 2×10^6 erg/cc. Differences in the square-shaped *M-H* loops were found among irradiated films, and the perpendicular remanence was decreased by increasing the ion irradiation dose, as shown in Fig. 3(b)-(d). It is assumed that a change in the

microstructure leads to the modification of the hysteresis loop after irradiation. The coercive force and the remanence ratio are very sensitive to aspects of the local structure of the multilayer film, such as defects, the grain size, and the periodic stacking structure. For ion doses below 5×10^{14} ions/cm², irradiated films had almost perfectly square perpendicular hysteresis loops, but the coercive force gradually decreased with increasing ion dose, and then abruptly decreased for films irradiated with an ion dose of 5×10^{14} ions/cm² at $H_c = 0.67$ kOe and $M_r/M_s = 0.83$, as shown in Fig. 4, and the magnetic anisotropy changed from the perpendicular to the in-plane direction at the highest ion dose, as shown in Fig. 3(d). The modification of the easy axis of the magnetic anisotropy following the ion irradiation is similar to the result previously reported by Chappert for ion-irradiated Co/Pt multilayer films⁹, and is thought to be responsible for the reduction of the interfacial anisotropy.

Fig. 3. Comparison of M - H loops for as-grown film (a), and irradiated films with various ion doses, 2×10^{13} (b), 5×10^{14} (c), and 5×10^{15} ions/cm² (d).

Fig. 4. Variation of the coercive field (▲) and remanence ratio (■) as a function of the Ga⁺ ion dose. The zero ion dose is for as-grown film.

Fig. 5. Variation of the effective magnetic anisotropy energy (K_{eff}) as a function of the Ga⁺ ion dose. The zero ion dose is for as-grown film.

The magnetic anisotropy of the Co/Pd multilayered film has been reported to be expressible as the sum of the interface anisotropy between Co and Pd layers and the crystalline and shape anisotropies of the Co layer. Carcia *et al.* introduced the following phenomenological equation involving the interface magnetic anisotropy of the Co/Pd layer and the volume anisotropy of the Co layer, $K_{eff} = K_v + 2K_s/t_{Co}$, where K_{eff} is the effective uniaxial magnetic anisotropy for the unit volume of the Co layer, K_v is the volume anisotropy of the Co layer, K_s is the interface magnetic anisotropy between the Co and Pd layers, and t_{Co} is the thickness of the Co layer¹⁰. Here, the value of K_v includes the contribution of the demagnetizing energy, and the positive values of K_{eff} , K_v , and K_s represent the perpendicular magnetic anisotropy. Fig. 5 shows the change in the effective magnetic anisotropy energy (K_{eff}) estimated from the amplitude of the torque curve for as-grown and irradiated films. In this investigation, the value of the effective magnetic anisotropy energy (K_{eff}) is given for a unit volume of the Co/Pd multilayer film. The interface anisotropy contribution, K_s , seems to dominate in as-grown film and is then reduced in irradiated films because of the interdiffusion of Co and Pd atoms in Co/Pd multilayer films. The change in the interface magnetic anisotropy caused by Ga ion irradiation is considered to be the main reason for the decrease in the effective magnetic anisotropy energy, K_{eff} , of Co/Pd multilayer films, because the magnetization, M_s , of as-grown films was almost constant after irradiation, as shown in Fig. 3. The effective perpendicular magnetic anisotropy energy, K_{eff} , was gradually decreased by increasing the Ga ion irradiation from 1.7×10^6 erg/cm³ for as-grown film to 0.4×10^6 erg/cm³ for film irradiated at 2×10^{15} ions/cm². The easy axis of magnetization changed from the perpendicular to the in-plane direction in film irradiated with an ion dose of around 5×10^{15} ions/cm², since the effective magnetic anisotropy energy became negative, as shown in Fig. 5. It is assumed that the ion-beam-induced atomic disordering of the interface during ion irradiation, as clarified in low-angle X-ray diffraction profiles, is responsible for the changing magnetic anisotropy of Co/Pd multilayer films.

However, the low-angle first peak of films irradiated with an ion dose of 5×10^{14} ions/cm² disappeared, but the effective magnetic anisotropy energy, K_{eff} , was still positive at 0.9×10^6 erg/cm³, indicating a perpendicular easy axis. Another possibility is that the interdiffusion of Co and Pd atoms in Co/Pd multilayer film after irradiation may change the Co/Pd structure into a CoPd/Pd structure, because the thickness of the Pd layer is much larger than that of the Co layer. Consequently, a low-angle first peak will be weakened in such a structure. But it should be noted that the effective magnetic anisotropy energy of CoPd/Pd multilayer structures was still positive¹⁴⁾. It is considered that multilayer structures still exist in that film. At an ion dose of 5×10^{14} ions/cm², ion irradiation not only enhanced the interdiffusion between the Co and Pd layers but also increased the roughness of the interface, resulting in a decrease of the low-angle diffraction peak.

Fig. 6. MFM image in the remanent state after application of an external magnetic field of 15 kOe along the surface normal direction: as-grown film (a), and after ion irradiation with 1×10^{14} ions/cm² (b), 5×10^{14} ions/cm² (c), 1×10^{15} ions/cm² (d), 2×10^{15} ions/cm² (e), and 5×10^{15} ions/cm² (f).

It has been stated that ion irradiation reduces perpendicular anisotropy, and it is expected that ion irradiation will also change the magnetic domain structure. The effects of ion irradiation on the magnetic domain structure were studied for as-grown and irradiated films. Fig. 6 shows typical MFM images of domain structures of as-grown and irradiated Co/Pd films in the remanent state. Here, the lift scan height during measurement was 50 nm. We magnetized as-grown and irradiated Co/Pd multilayer films once by applying a strong external magnetic field (15 kOe) in the direction normal to the film plane, and obtained MFM images in a zero field. The MFM tip was magnetized in the *z* direction (perpendicular to the plane of the sample) prior to the measurement, and the magnetic contrast can be explained by an interaction between the perpendicular magnetized Co/Pd multilayers and the MFM tip. As-grown film had a uniform contrast, as shown in Fig. 6(a). This result suggests that the remanent state of as-grown film can be described as a single magnetic domain, because it has large remanent magnetization almost equal to the saturation magnetization, as shown in Fig. 3(a). In film irradiated with an ion dose of 1×10^{14} ions/cm² (Fig. 6(b)), some bright spots were clearly seen, which may reflect a transition from a single-domain into a maze-domain structure. The maze-domain structure was clearly seen in irradiated films, as shown in Fig. 6(c)-(f). In irradiated films, the typical maze-domain structure still had a similar irregular appearance when the ion irradiation dose was increased, but the magnetic domain size decreased. It has been noted that width and separation of the domain structures is determined by the ratio of the magnetic wall energy $\sqrt{2A_0K_{\text{eff}}}$ to the demagnetization energy, $2\pi M_s^2$, where A_0 is the exchange constant, K_{eff} is the effective magnetic anisotropy energy, and M_s is the saturation magnetization¹⁵⁾. A substantial decrease in magnetic domain size was evident when the ion irradiation dose was increased. Since the magnetization M_s of as-grown and irradiated films was almost the same, a decrease in the magnetic domain size is due to a decrease in the domain wall energy, which corresponds to a decrease in the effective magnetic anisotropy, as confirmed in Fig. 5. The contrast decreased with increasing ion irradiation doses and with the resulting decrease in the domain size. In films irradiated with ion doses of 2×10^{15} ions/cm² and 5×10^{15} ions/cm², the contrast decreased especially significantly. This might be due to a decrease in the size of the perpendicularly magnetized domains.

From the observed stripe domain width of ~ 200 nm for the sample dosed at 1×10^{15} ions/cm², as shown in Fig. 6(d), the domain wall energy was estimated to be ~ 6 erg/cm² for this sample, assuming the demagnetizing energy of parallel band domains in a thin perpendicular magnetized film¹⁶⁾. The uniaxial anisotropy K_u (which is $K_{\text{eff}} + 2\pi M_s^2$) was 2.1×10^6 erg/cc. Thus, by assuming the simple expression $4\sqrt{A_0K_u}$ for the energy density of Bloch walls and neglecting the demagnetizing energy in the wall, the exchange stiffness A_0 is estimated to be $1.1 \times$

10^{-6} erg/cm. If we use these values and assume the simple expression $\pi\sqrt{A_0/K_u}$ for the domain wall width, the wall width can be estimated at ~ 20 nm for a sample irradiated with an ion dose of 1×10^{15} ions/cm². The estimated domain walls are considered to give a measure of the space (transitional space) between patterned bits in planar patterned Co/Pd media.

Fig. 7. AFM images of an array of squares 1 μm in size with a spacing of 0.7 μm : plane view (a) and 3-dimensional view (b).

Fig. 8. MFM image of ion-irradiation-patterned films with a pattern size of 1 μm and a spacing of 0.7 μm .

3.2 Ion-irradiation-patterned Co/Pd multilayers

Fig. 7 shows AFM images of ion-irradiation-patterned Co/Pd multilayer films irradiated at an ion dose of 5×10^{15} ions/cm². The etching depth of the irradiated region is about 4.5 nm. The ion-irradiation-patterned Co/Pd film was smooth, showing regions of 10 $\mu\text{m} \times 10 \mu\text{m}$, where squares with a size of 1 $\mu\text{m} \times 1 \mu\text{m}$ were separated by a distance of 0.7 μm , as shown in Fig. 7(a). In addition, the surface roughness of ion-irradiation-patterned film is an important property. Three-dimensional AFM images of patterned media give further insight into modification at the surfaces, as shown in Fig. 7(b). An MFM image of a patterned film is shown in Fig. 8. This image was taken without any external magnetic field applied after the sample deposition. The significant difference in magnetic contrast and typical domain structures between un-irradiated and irradiated regions can be clearly observed in the patterned film. The MFM image of the patterned film indicates that each un-irradiated square consists of localized maze domain structures, which corresponds to perpendicular magnetization direction. Low magnetic contrast and the absence of maze structures in irradiated areas may be due to the change of the magnetization direction from perpendicular to in-plane. The boundary between the perpendicular region and in-plane region is very sharp. It is considered that the Ga ion beam induces atomic disordering and separates the magnetized regions, which may be useful for separating bits on media.

4. Conclusion

We have demonstrated that Ga ion irradiation modified the magnetic properties and microstructures of Co/Pd multilayer films. The effect of an ion dose on the layered structure was confirmed by low-angle XRD measurement. The first peak due to the multilayer structure disappeared after irradiation with an ion dose of 5×10^{14} ions/cm². Ion irradiation had a significant influence on the regularity of the periodic structure in Co/Pd multilayer films because of the interdiffusion between Co and Pd atoms at the interface. The modifications of magnetic properties by Ga ion irradiations were associated with a decrease in the interfacial magnetic anisotropy. At lower Ga ion doses ($< 5 \times 10^{14}$ ions/cm²), the multilayer film retained its perpendicular anisotropy, but the coercive force gradually decreased. The transition from perpendicular to in-plane direction occurred at a Ga ion irradiation dose of around 2×10^{15} ions/cm². A further decrease in the perpendicular magnetic anisotropy was observed at higher Ga ion doses. MFM images of the irradiated films showed that the domain width gradually decreased with increasing ion dose. Significant differences in magnetic contrast and domain structure between irradiated and un-irradiated regions were observed in the patterned films. A sharp boundary between perpendicular and in-plane regions was also observed, indicating that localized patterns can be engineered by Ga ion irradiation.

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