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Influence of an Fe cap layer on the structural and magnetic properties of $Fe_{49}Pt_{51}$/Fe bi-layers*

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The influences of an Fe cap layer on the structural and magnetic properties of FePt/Fe bi-layers are investigated. Compared with single FePt alloy films, a thin Fe layer can affect the crystalline orientation and improve the chemical ordering of $L1_0$ FePt films. Moreover, the coercivity increases when a thin Fe layer covers the FePt layer. Beyond a critical thickness, however, the Fe cover layer quickens the magnetization reversal of $Fe_{49}Pt_{51}$/Fe bi-layers by their exchange coupling.

Keywords: Fe cap layer, $L1_0$ FePt, magnetism
PACC: 7550, 7560G

1. Introduction

The ordered face-centered tetragonal (fct) $L1_0$ FePt shows a large magneto-crystalline anisotropy ($K_u \sim 7 \times 10^7$ erg/cc), which enables the reduction of recording bite size and delays the onset of superparamagnetism. Therefore, the $L1_0$ FePt is considered to be promising as a future ultrahigh density recording media (1 Tbit/in^2). However, a disordered fcc (A1) structure usually forms in the as-deposited FePt thin film prepared at room temperature. Then a phase transformation from A1 to $L1_0$ is necessary to achieve hard magnetic properties. $Ex-situ$ annealing at high temperature will cause the fast growth of magnetic grains,[1] resulting in large surface roughness and high noise. Thus, it is very important to find a way to fabricate the $L1_0$ FePt film at relatively low temperature.

To date, many methods have been reported to obtain $L1_0$ FePt- based films at a temperature lower than 600°C, such as the introduction of under-layers,[2−6] the addition of the third elements,[7−10] multi-layering,[11−13] and monatomic layer deposition.[14 It is proved to be an efficient way to promote the chemical ordering of FePt films by the addition of Cu, Ag, B, etc. Temperature reduction by Cu addition is ascribed to the increased driving force arising from the formation of the ordered FePtCu alloy,[7] or decrease of the melting temperature, which in turn enhances the atom diffusivity.[8] However, Barmak *et al* asserted that the equivalent addition of Fe has the same effect on lowering the kinetic ordering temperature.[9] As a result, the off-stoichiometric, Fe-rich FePt film is a suitable candidate for recording media.[6,15]

It is interesting to know the influence of the Fe cap layer on the structural and magnetic properties. Considering the possible diffusion in the interface, the FePt composition changes near the interface, and an Fe-rich sub-layer will form. According to the results of Barmak, the sub-layer is assumed to order first at relative low temperature, and then promote the ordering of the rest of FePt film since the chemical ordering is an exothermic process. In this paper, the $Fe_{49}Pt_{51}$ (15 nm)/Fe films with a variable thickness, as well as FePt films with different Fe content, are prepared to investigate the influence of the Fe cap layer on the structural and magnetic properties of FePt/Fe bi-layers.

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2. Experiment

Thin film samples with the structure Glass/FePt/Fe were fabricated in a dc magnetron sputtering system. The background was better than 5.0×10^{-8} Torr (1 Torr=133 Pa). The working gas pressure of high-purity argon (99.99%) was fixed at 5 mTorr. First, an FePt layer with a nominal thickness of 15 nm was prepared on corning glass substrates by co-sputtering at an $in-situ$ temperature of 400 °C. Except for special indication, the sputtering power for Fe and Pt targets was fixed at 70 W and 40 W, respectively. Then Fe layers, with nominal thickness of 0.8, 1.7, 3.4 and 6.8 nm were deposited at the same temperature just after the deposition of the FePt layer, and the sputtering power was fixed at 80 W. The temperature kept for another 10 minutes after deposition. As a comparison, the samples with a similar structure Glass/FePt/Ag were fabricated by the same process. And the samples with different Fe contents were fabricated just by changing the sputtering power of Fe target (from 50 W to 120 W), meanwhile keeping the power of Pt. Finally, all the samples were annealed at 500 °C for 30 minutes.

The crystallographic structure of the samples was examined by x-ray diffraction (XRD) with Cu K_α radiation. Usually, the chemical ordering parameter S was calculated from the ratio of the integrated intensities of the (001) to (002) peaks derived from the XRD pattern.[11,14,15] In our case, however, films did not have a good texture. Moreover, the intensity of the (001) peak was hard to estimate because of the diffraction of the glass substrate. So, the following formula was used:[16]

$$S^2 = (1 - c/a)/[1 - (c/a)_{S_f}], \qquad (1)$$

where $(c/a)_{S_f}$ was the axial ratio for the fully ordered phase (S_f), and c/a for the partially ordered phase. $(c/a)_{S_f}$ was determined to be 0.964 according to the powder diffraction data in JCPDS card (PDF No. 43-1359). Magnetic properties of the samples were measured with a vibrating sample magnetometer (VSM) at room temperature and Super-conduct Quantum Interfere Devices (SQUIDs) at 100 K. The composition of the FePt films was measured by inductively coupled plasma-mass spectrometers (ICP-MS) and x-ray fluorescence (XRF). And the composition analysis along the cross section was performed by Auger electron spectrum (AES).

3. Results and discussion

Figure 1 shows the XRD θ–2θ spectra of FePt alloy films annealed at 500 °C. The Fe content increases from 45.6 at% (A) to 61 at% (F). First, all the alloy films exhibit (111) texture, and the (111) peak migrates to high angle gradually as the Fe content increases. The migration indicates the contraction of the crystal lattice. There are two reasons. The main reason is that the decrease of plane spacing, due to the reduced c-axis of the $L1_0$ phase, leads to the migration of the (111) peaks. Furthermore, at the condition of high Fe content, substituted Fe atoms where radium is smaller than that of Pt, keep the trend. Next, there is a very weak FePt (001) super-lattice peak in the films, indicating they are ordered. Furthermore, the (200) and the (002) peak are too faint to observe. The chemical ordering parameter cannot be calculated as the ratio of intensity of (001) peak to that of the (002) one. Finally, an Fe (110) peak is found in film F with the nominal composition of $Fe_{61}Pt_{39}$. Fe particles form in the film.

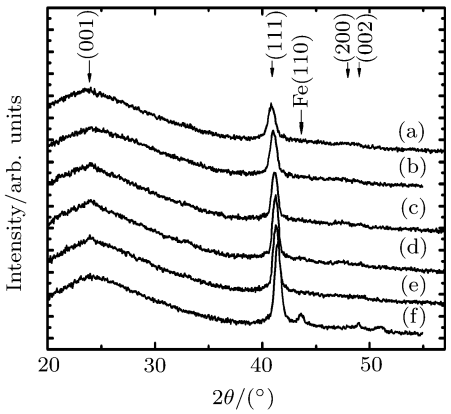


Fig.1. The XRD θ–2θ spectra of co-sputtering FePt alloy films. The Fe content is below: (a) 45.6 at%, (b) 49 at%, (c) 52 at%, (d) 54.7 at%, (e) 57 at%, (f) 61 at%. All the films are annealed at 500 °C for 30 min.

Figure 2 exhibits the XRD spectra of $Fe_{49}Pt_{51}$ films covered by an Fe cap layer. Except for FePt (111), an obvious (001) super-lattice peak, as well as the (200) and (002) peaks, can be seen in the bilayer with a 0.8-nm-thick Fe. The (001) peak becomes very strong when the thickness of the Fe layer increases up to 1.7 nm in (c). Its intensity is larger than that of the (111) peak. Normally, the intensity of the (001) peak is about a third of that of the (111) peak in the powder diffraction. So, the (001) orientation has been greatly improved by covering a thin Fe layer. Simultaneously, the chemical ordering parameter S,

evaluated by the change of lattice constant, is 0.98, which indicates that the film is almost full ordered. In addition, the (200) peak disappears. With further increase of the Fe layer, (001) and (002) peaks turn weak as shown in (d). At last, an Fe (110) peak appears when the thickness of Fe reaches 6.8 nm. It is well known that there is a re-crystallization process during annealing.[17] The above results indicate that the re-crystallization of FePt films with an Fe cap layer is much different from that of co-sputtering films, and that Fe layers can be used to improve the film orientation.

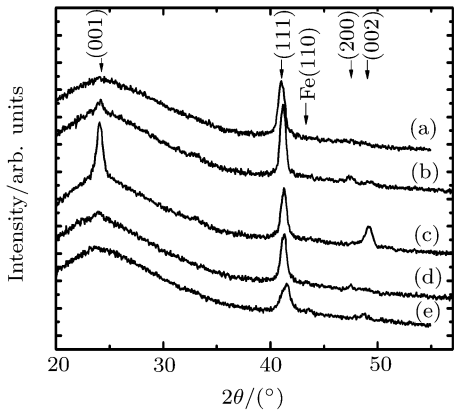


Fig.2. The XRD θ–2θ spectra of $Fe_{49}Pt_{51}$/Fe bilayer films. The thickness of the Fe cap layer is: (a) 0, (b) 0.8 nm, (c) 1.7 nm, (d) 3.4 nm, and (e) 6.8 nm. All the films are annealed at 500 °C for 30 min.

Figure 3 shows the dependence of in-plane coercivity of Fe-Pt alloy films on the Fe content prepared by the co-sputtering method. The coercivity of annealed alloy films depends strongly on the film composition. It is 2.3 kOe (1 Oe=80 A/m) when the Fe content is 45.6 at%. Then it increases quickly when the Fe content increases, and reaches the maximum $H_C = 15.5$ kOe in the film with the Fe content 52 at% at the condition of Fe power of 80 W. The further increase of Fe power results in a drastic decrease of coercivity. The maximum is about six times larger than the minimum. It is well known that coercivity is an extrinsic characteristic, decided not only by the intrinsic ones such as anisotropy and magnetization, but also by the structural properties of the samples. As shown in figure 1, all the alloy films have similar film orientation, and the grain sizes calculated by the Scherrer formula are nearly the same. Then the coercivity dependence should be interpreted as the magneto-crystalline anisotropy dependence on the composition. It is clear that the anisotropy is in proportion to the chemical ordering parameter of the FePt film, and varies with the film composition.[18] Normally, the magnetic property of FePt films changes from soft magnetism of disorder FCC phase to the hard one of the $L1_0$ phase. The coercivity change is great. In contrast, the coercivity variation is relatively small with the film composition.[18] As a result, the remarkable change of coercivity is mainly due to the difference of chemical ordering. That is, the highest ordering degree has been obtained in the film with Fe content around 52 at% after 500 °C annealing. On the other hand, the films with high (61.1 at%) or low (45.6 at%) Fe content are difficult to order at the same conditions. A higher temperature is needed for these films. The results are the same for FePt alloy films prepared at the substrate temperature of 400 °C, and the coercivity increases three times with the composition. In brief, a little excess Fe is helpful for the chemical ordering of FePt films at the same temperature. It coincides with another group's work.[10]

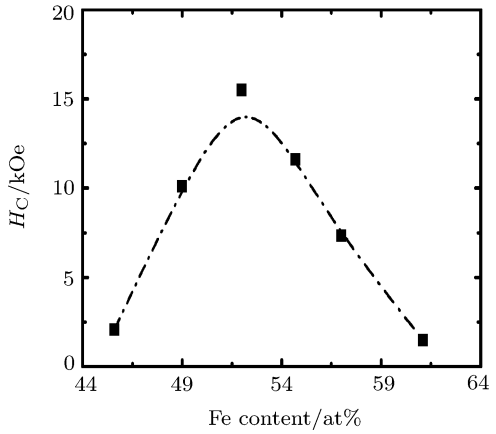


Fig.3. The coercivity dependence of co-sputtering FePt alloy films on the Fe content.

Figure 4 shows the dependence of in-plane coercivity of $Fe_{49}Pt_{51}$/Fe bi-layers on the thickness of Fe cap layers. The coercivity increases with the thickness of the Fe cap layer, reaches a maximum 17.5 kOe when the thickness is 0.8 nm, and then decreases with further increase of the thickness. Meanwhile, the coercivity of the bilayer with a 1.7 nm Fe top layer is still higher than the single FePt film. In general, the $Fe_{49}Pt_{51}$/Fe bi-layers can be regarded as an exchange-spring magnet, consisting of a hard layer and a soft one. This kind of structure is favorable to increase the magnetic energy product, but results in the coercivity decrease which depends strongly on the thickness of the soft layer.[19] In the current case, however, the coercivity increases nearly twice as much when a thin

Fe layer covers an FePt film. Therefore, evolutions should be expected. First of all, there is a change of the film orientation. Second, vacancies may induce by the bombardment of Fe atoms.[4] Third, the chemical ordering of the FePt film has improved. Finally, there may be atoms diffusion between the two layers during 500 °C annealing, leading to composition variation of the FePt and promoting the ordering. Compared to the influence of chemical ordering of FePt films, that of orientation change is very small on the coercivity variation.[13] So, the minor structural change of $Fe_{49}Pt_{51}$/Fe (0.8 nm) shown in figure 2(b) from the $Fe_{49}Pt_{51}$ film should not be the reason leading to the drastic change of coercivity. The influence of vacancies is assumed to be small because Fe is not the third element. And, the addition of an Ag top layer decouples the FePt grains, resulting in the coercivity enhancement.[4] Furthermore, the $Fe_{49}Pt_{51}$ layer in the bilayer film is prepared under the same condition and has the same composition as those of the $Fe_{49}Pt_{51}$ film. They would have the similar structural and magnetic properties. As discussed above, however, the film composition will affect the chemical ordering of the FePt film, and high ordering is obtained in the film with a little more Fe. Then diffusion may be the key point to understand the coercivity change. Usually, the thickness of a single Fe or Pt layer is no more than 3 nm in the multi-layers deposition.[2,10−13] The diffusion length decreases in an exponential decay along the depth. The thicker the film is, the more difficult the uniformity is to obtain. Moreover, the uneven composition distribution along depth has been qualitatively revealed in the Fe/FePt bilayer. Fe content in the interface between FePt and Fe is richer than that in the bottom of FePt film. As a consequence, a reasonable explanation is that the diffusion leads to a composition change in the FePt film, and an Fe-rich FePt sub-layer with the composition 52:48 will order first, and simultaneously give out thermal energy which promotes the ordering of the rest[20], so that a highly ordered state is achieved.

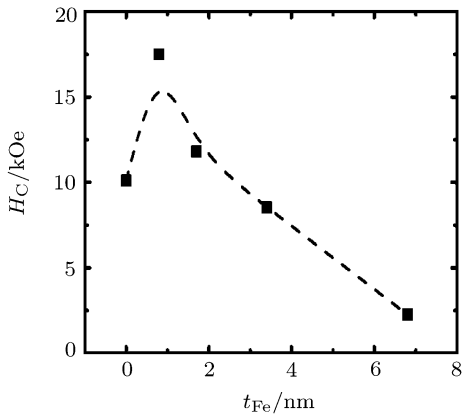


Fig.4. The coercivity dependence of $Fe_{49}Pt_{51}$/Fe bilayer films on the thickness of the Fe cap layer.

The coercivity of the $Fe_{49}Pt_{51}$/Fe bi-layers with a 1.7 nm Fe layer is much smaller than that with a 0.8 nm Fe layer, although it has a high ordering parameter of 0.98; and the coercivity decreases greatly with the further increase of Fe layer thickness. This implies that there is a diffusion-free Fe layer which acts as the anti-magnetization nuclei of the magnetization reversal. In other words, there is a critical thickness of diffusion between 0.8 nm and 1.7 nm. Beyond the critical one, the magnetization reversal can be explained by the model of an exchange-spring magnet.

Figure 5 shows the loops of $Fe_{52}Pt_{48}$ and $Fe_{61}Pt_{39}$ films. For the $Fe_{52}Pt_{48}$film, the easy axis closes to the film plane. It is reasonable because the angle between the magnetic easy axis and the normal direction is about 54° in the case of a (111) texture. For $Fe_{61}Pt_{39}$ film, however, the easy axis is in the plane and its anisotropy decreases greatly.

For comparison, the hysteresis loops of $Fe_{49}Pt_{51}$/Fe bi-layers are indicated in figure 6. The thickness of the Fe layers are 0.8 nm and 6.8 nm, respectively. The Fe content in the bi-layers with a 0.8 nm Fe is 51.7 at%, very similar to that of $Fe_{52}Pt_{48}$co-sputtering films. After the film is covered by a 0.8 nm Fe layer, both the in-plane and out-of-plane coercivity increase and the easy axis tends to a normal direction. This confirms that both the structural and magnetic properties are changing. However, the bilayer with a 6.8 nm Fe cap layer is a typical exchange-spring magnet in which the soft and hard layers couple together, because the thickness of the Fe layer is smaller than the exchange coupling length. As a result, they will reversal as a whole and the soft layer acts as anti-magnetization nuclei. Then the coercivity decrease greatly as shown in figure 6(c). Nevertheless, there is a trail of the hard magnetic layer. The saturation field is about 10 kOe, although the coercivity is no more than 2.3 kOe. Furthermore, the out-of-plane loop indicates the different magnetization reversal of the Fe and $L1_0$ FePt layer. The coercivity of the hard layer is no less than 8.5 kOe.

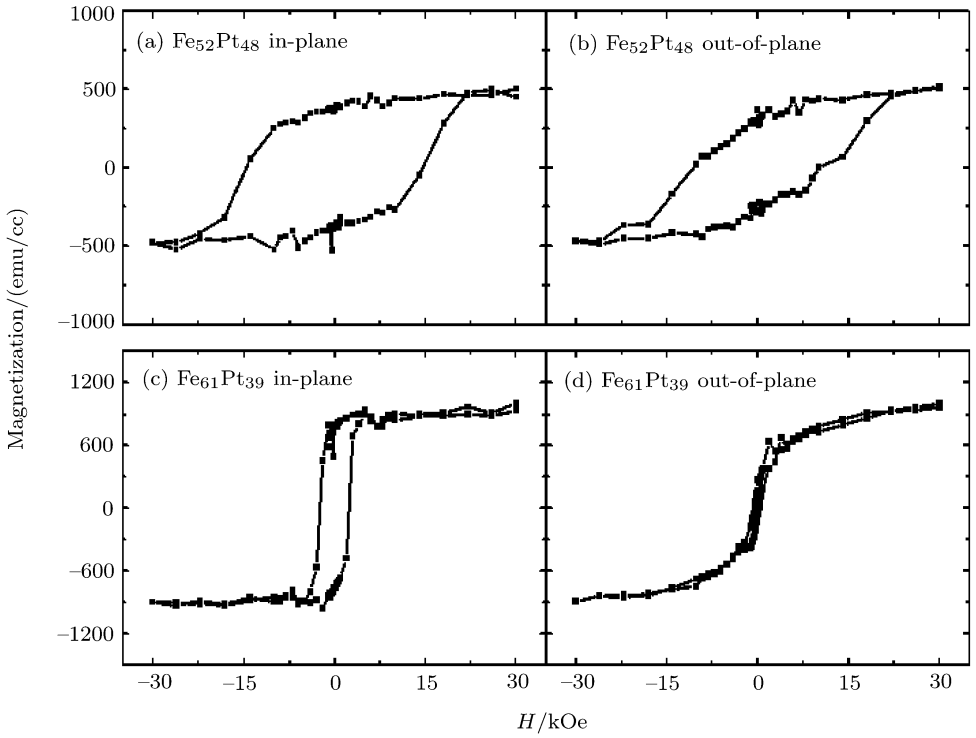


Fig.5. The in-plane and out-of-plane magnetic hysteresis loops of $Fe_{52}Pt_{48}$ and $Fe_{61}Pt_{39}$ films.

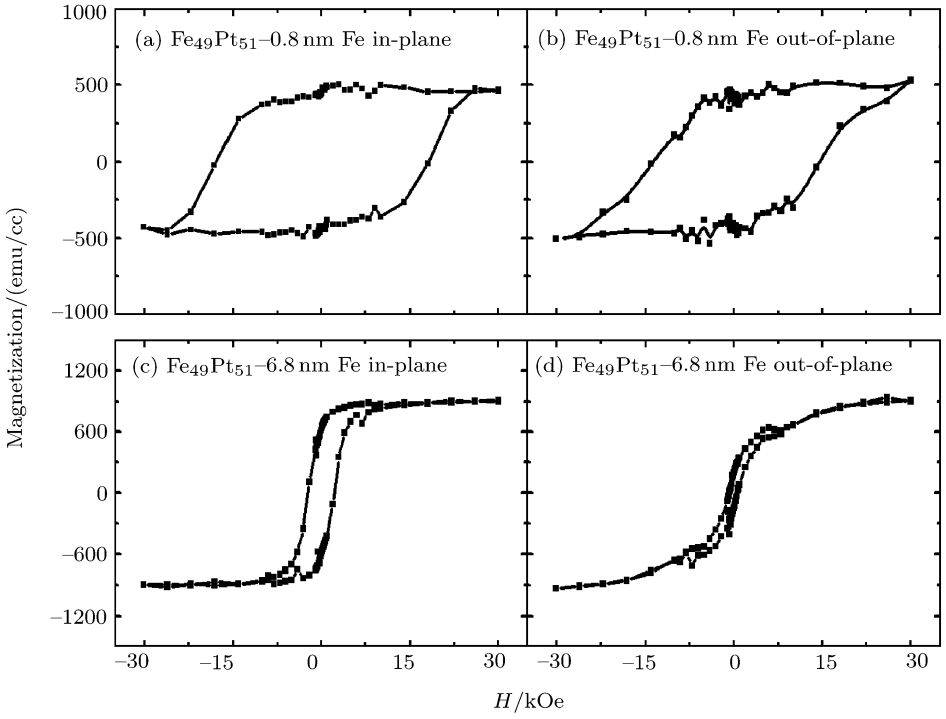


Fig.6. The in-plane and out-of-plane magnetic hysteresis loops of $Fe_{49}Pt_{51}$/Fe bi-layers with Fe layers of thicknesses 0.8 nm and 6.8 nm, respectively.

4. Conclusion

The structural and magnetic properties of $Fe_{49}Pt_{51}$ films with Fe cap layers are investigated. The high ordered (001) textured FePt film is obtained in the case of a 1.7 nm Fe cap layer. At the same time, the diffusion

of Fe atoms in the interface between Fe and FePt layers results in the composition change which promotes the chemical ordering of FePt films. Therefore, the coercivity increases while a thin Fe layer covers on the FePt layer. When the Fe thickness exceeds a critical value, however, the residual Fe layer acts as the nuclei of the magnetization reversal, and then the coercivity of FePt/Fe bi-layers is reduced greatly with the increase of Fe layer thickness.

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