Fine control of $L1_0$ ordering and grain growth kinetics by C doping in FePt films

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We present the results on highly anisotropic oriented face-centered-tetragonal ordered equiatomic FePt thin films embedded in a C matrix prepared by cosputtering technique on MgO (100) substrates at 400 °C. We have found that doping of C in FePt alloy films is an effective and appropriate way to control the $L1_0$ ordering and growth of grain size down to 4 nm. Room-temperature magnetic measurements reveal that perpendicular magnetic anisotropy exists for these films with C concentration up to 25 vol.%. These films are very promising and the results are of relevance for ultrahigh-density magnetic recording media. © 2003 American Institute of Physics.

Equiatomic FePt alloy thin films have received significant attention as possible magneto-optic recording media and high-energy permanent magnets owing mainly to the existence of ordered intermetallic magnetic phases with exceptional magnetic properties such as large uniaxial magnetic anisotropy energy and enhanced magneto-optical Kerr rotation. Moreover, in the search for perpendicular magnetic recording media with high anisotropy, high coercivity, and low media noise, these alloys have shown promising characteristics.1–7 However, in these materials a high temperature treatment (substrate heating during deposition and/or postannealing) is necessary to obtain the ordered phase with high values of magnetocrystalline anisotropy.8 Recent studies on microstructural properties of FePt thin films7 and on FePt nanoparticle9 assemblies reveal that the magnetic grains in the FePt system are interconnected to some degree. As a result, it is expected that the grains will be magnetically coupled between each other and the reduction of interparticle distances with increasing substrate/annealing temperature results in dramatic change in the interparticle interactions.

On the other hand, ultrahigh-density magnetic recording with low media noise imposes the need of a material, which consists of magnetically isolated grains with size below 10 nm.10 Moreover, in such small-size isolated particles, large magnetic anisotropy is required to overcome the thermal fluctuation and demagnetization fields. So, in order to control grain size of the magnetic particles as well as to achieve magnetic isolation among grains, various substitutions of nonmagnetic elements (in multilayer fashion, introduction of buffer layer, and/or exchange-decoupling layer between the FePt layer and soft-magnetic under layer) have been investigated11–15 in the FePt system. This substitution obviously determines the mechanism of grain growth, interparticle separation, i.e., magnetic isolation and therefore, the interparticle interactions.16 However, most of the studies on the earlier systems are followed by annealing the (001) oriented multilayer precursors and obtaining the magnetic properties superior to those of the as-deposited films.13,17 Although this process results in films with desired magnetic properties, it is observed to be somewhat complicated, which involve both multilayer deposition and postdeposition annealing. Moreover, this method has disadvantages as a manufacturing process because such a high temperature postdeposition annealing can cause irreversible changes in substrates for magnetic media and enhances the particle growth considerably.

So, in this letter we report a further simplified fabrication method, direct synthesis of $c$-axis oriented ordered (FePt)$_{1-x}$C$_x$ by cosputtering at 400 °C substrate temperature, with resulting properties of a mean grain size of 5.2 nm and perpendicular coercivity of about 5 kOe, showing an improvement over the previous studies.13,15 For the purpose of forming small grains of FePt with weak exchange coupling, C would be an ideal material because there exists neither stable Fe nor Pt carbides. Structural evidence establishing the formation of ordering phases with varying degrees of chemical order and grain growth kinetics are presented, together with their correlation with the resulting magnetic properties.

50-nm-thick equiatomic FePt and FePt:C films were prepared directly on MgO (100) substrates using dc magnetron cosputtering of composite FePt and C targets with a base pressure of better than $8 \times 10^{-7}$ Torr and the sputtering Ar gas pressure of 5 mTorr. A composite FePt target was made by putting some Pt (99.99% purity) chips on the Fe target (99.99% purity). The volume concentration of the C content ($x$) is varied up to 50%. In the case of the FePt thin films, the samples were prepared at different substrate temperatures varying from room temperature to 500 °C by using home-built and calibrated heater setup, while in the FePt:C system the samples were prepared at constant substrate temperature of 400 °C with various amounts of C doping. The crystal structure of the films was characterized by Rigaku D/MAX-RC and RB x-ray diffractometer using Cu $K\alpha$ radiation. The microstructure and surface morphology of the films were investigated by transmission electron microscopy (CM20T) and scanning electron microscopy (Philips XL30SFEG), respectively. The atomic ratio of Fe/Pt of all the films prepared in this investigation was found to be an
almost constant ratio of 49.5:50.5, measured by means of an energy dispersive x-ray analysis. Room-temperature magnetic characterizations were carried out by a vibrating sample magnetometer (VSM) with an applied field of up to ±20 kOe and a torque magnetometer with an applied field of 10 kOe.

Figure 1 shows the x-ray diffraction (XRD) pattern for 50 nm equiatomic FePt samples prepared at different substrate temperatures. The sample prepared at room temperature does not show any film orientation peaks. On the other hand, for the sample prepared at 150 °C shows only one peak at about 48.3°, which corresponds to neither (200) (2θ = 47.6°) nor (002) (2θ = 49.3°) of the face-centered-tetragonal (fct) ordered FePt, but rather the position of the peak is in-between. The hysteresis loop shows very small coercivity for this sample, indicating that the sample structure is a disordered fcc phase. Upon increasing the substrate temperature, the peak observed at 48.3° broadens at initial stages and ultimately splits into two peaks corresponding to (200) and (002) peaks of the ordered fct phase. The splitting of peaks could be seen clearly in XRD data for the sample prepared at 300 °C substrate temperature. As the substrate temperature increases above 200 °C, the (001) peak (characteristic of $L1_0$ chemical ordering) becomes stronger and dominates at 500 °C, while the (002) peak dominates over the (200) peak above 300 °C, showing that the film is becoming more dominated by a c-axis normal texture. The detailed investigation of surface morphology of the FePt samples by means of scanning electron microscopy shows the island-like structure with an average island size of 170 nm. The in-depth investigation of the island structure by high-resolution transmission electron microscopy showed that the islands have granular structure with FePt grains of an average size 5–10 nm and the ordered $L1_0$ structure was developed inside the FePt grains. However, these grains are not completely isolated with each other and yield strong interparticle exchange interactions.

In order to control the grain growth kinetics and $L1_0$ ordering in FePt films, we have doped various amount of C in FePt thin films at 400 °C substrate temperature. The choice of the sample, prepared at 400 °C substrate temperature, has been dictated by the fact that the sample has a remanence ratio of almost 100% in room-temperature hysteresis loop as demonstrated later and the average grain size of 15–50 nm. The sample of $(\text{FePt})_{1-x}\text{C}_x$ nanocomposite thin films deposited on MgO (100) substrates, consists of nanometer size, $L1_0$ structured FePt particles embedded in a C matrix. Figure 2 shows the XRD pattern of FePt:C films prepared at 400 °C substrate temperature with various amounts of C doping. Except the peak from the MgO substrate, all visible peaks are attributed to the fcc or fct FePt phase, indicating that most C remains as a pure element matrix rather than forming any carbide phases. It is clear from the figure that the intensity of the $L1_0$ ordering peaks is predominant initially, then reduced with increasing C doping. Subsequently, the ratio of the disordered and ordered FePt (200)/(002) peaks increases with increasing C addition and merge into a single peak for 50% C doping, suggesting that the amount of ordered and disordered materials in FePt system could be controlled adequately by C doping. The average grain size calculated by Scherrer’s formula is found to decrease with C doping and attains a minimum value of about 3 nm for 50% C-doping films. Moreover, the order parameter calculated from the intensity of the superlattice peaks versus the fundamental peak is also found to decrease with C doping.

The microstructure of the FePt:C films was investigated by transmission electron microscopy (TEM). Figure 3 shows the microstructure of the C-doped FePt thin films together with undoped FePt film. It is important to note that the particle size found in FePt system is larger in size and has a wide range from 15 to 30 nm. As the C doping increases, the particle size noticeably decreases compared to that of pure FePt system. This might be due to the preventing of growth of the FePt particles by C when a small amount of C is doped and the regular arrangements and isolation of FePt particles takes place due to agglomeration at grain boundaries with more C addition. The electrical transport measurement reveals that there is a one order of magnitude change in the electrical resistance between 25% C (20 Ω) and 33% C (330 Ω) doping. The mean particle size for the film with 25% C doping, obtained from the TEM microstructure and calculated from the corresponding histogram, is 5.2 nm and in the case of 50% C doping, the particles size is further reduced to 3.7 nm.
pure FePt system to 1.08 $L_1_0$ ordering is due primarily to the decrease in anisotropy energy decreases from 2.82 ergs/cm$^3$ for the FePt system. The determined value of magnetic anisotropy ratio was observed as a function of C reorientation transition from perpendicular magnetic anisotropy with lower C doping, suggesting that this system might be applicable to accomplish a magnetic recording medium capable of storage density beyond 1 Tb/in$^2$.

In conclusion, FePt:C nanocomposite thin films with strong perpendicular magnetic anisotropy were fabricated by using cosputtering technique directly on a MgO($100$) substrate at elevated substrate temperatures. Under the appropriate C doping, these films consist of $L_1_0$ FePt particles embedded in the C matrix with c axes of the grains oriented along the film normal direction. These films have fine grains (<7 nm), adjustable coercivity, $L_1_0$ ordering, and perpendicular magnetic anisotropy with lower C doping, suggesting that this system might be applicable to accomplish a magnetic recording medium capable of storage density beyond 1 Tb/in$^2$.

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FIG. 3. Transmission electron micrographs of the (FePt)$_x$C$_{1-x}$ system prepared at 400 °C substrate temperature with C doping of (a) 0%, (b) 25%, and (c) 50% and their grain size distributions. The horizontal bar in the TEM picture scales of 20 nm.

4 nm. These results are in close agreement to the XRD data within the experimental error. Although the substrates and preparation techniques were different from the previous study, the results obtained in the present investigated samples show a better improvement (the mean grain size of 5.2 nm and perpendicular coercivity of about 5 kOe) than earlier report, where the mean grain size is shown to be 8 nm after optimum annealing at 700 °C.

The magnetic properties of the samples were characterized using a torque magnetometer and VSM. The magnetic anisotropy energy and the easy axis orientation were determined by a simple analytical method from a measured torque curve. The easy axis of the samples with C concentration up to 25% is close to 90°, suggests that the easy axis is normal to the film plane, while the 50% C-doped sample shows the easy axis parallel to the film plane. A clear spin-reorientation transition from perpendicular magnetic anisotropy to in-plane anisotropy was observed as a function of C doping in the FePt system. The determined value of magnetic anisotropy energy decreases from 2.82×10$^7$ ergs/cm$^3$ for the pure FePt system to 1.08×10$^7$ ergs/cm$^3$ for 75% FePt in the FePt:C sample. The subsequent decrease of anisotropy with C doping is due primarily to the decrease of complete $L_1_0$ ordering, which could be seen clearly from the XRD, where the intensity ratio of the predominant ordering peaks and the ratio of the $L_1_0$ superstructure peaks versus fundamental peaks decreases. Hence, our results support that the magnetic anisotropy in this system is mainly ascribed to magnetic-crystalline anisotropy along (002) oriented fct phase, similarly as reported in the FePt system. The $M$–$H$ hysteresis loop measured with the maximum field of 20 kOe applied normal to the film plane shows a remanence close to 100% of the saturation magnetization ($M_s$ = 1010 emu/cm$^3$) for the FePt film and decreases to 62% of its saturation magnetization ($M_s$ = 195 emu/cm$^3$) for the 50% C-doping film. This is close to the remanence value of 50% of the saturation magnetization expected for noninteracting and randomly oriented uniaxial grains.