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Structural and magnetic properties of nanocomposite Co:C films

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The structural and magnetic properties of Co:C nanocomposite films at room temperature were investigated as functions of annealing temperature, Co concentration, and film thickness. The as-deposited films, which were cosputtered from Co and C onto water-cooled glass substrates, are nonmagnetic amorphous Co–C alloys. Hexagonal-close-packed (hcp) Co grains are formed in 100 nm films annealed at 300 °C and most Co takes this structure when annealed at higher temperatures. The sizes of the Co grains range from 10 to 25 nm, with larger grain size resulting from higher annealing temperature and higher Co concentration. With the increase of annealing temperature and Co concentration, the magnetic activation or switching volumes increase faster than the physical grain volumes, suggesting the increased exchange coupling between neighboring Co grains. The films with high coercivities have optimal combinations of large grain size and weak intergrain exchange coupling before a network-like structure is formed and the percolation threshold is reached. Coercivities of about 800 Oe were obtained in 100 nm films with annealing temperature of 400 °C and Co concentration of 60 at. %. Higher coercivities, up to 1030 Oe, were obtained in films with reduced thickness and elevated annealing temperature. © 1999 American Institute of Physics. [S0021-8979(99)50708-1]

I. INTRODUCTION

The magnetic behavior of nanocomposite films has been extensively studied for their potential applications in magnetic recording¹ and magnetoresistance effect.^{2,3} Co and C are immiscible and the metastable Co carbides (Co₂C and Co₃C) decompose easily into Co and C.⁴ In fact the macroscopic interdiffusion coefficient between Co and C is negative.⁵ Co:C composite films therefore form a good system to investigate the magnetic properties of nanostructured Co particles. Hayashi *et al.* proposed this system for potential ultrahigh density magnetic recording since C can provide good isolation between neighboring Co particles to reduce the intergrain exchange interaction.⁶

Activation volume (or switching volume) V^* is the unit volume of magnetic moments switching together in magnetization reversal. For Stoner–Wohlfarth particles (single-domain particle with uniaxial anisotropy and coherent rotation) with volume V , information on the intergrain exchange interaction can be inferred from the comparison between V^* and V under low field approximation ($H_c \ll H_A$, where H_c is the coercivity and H_A is the uniaxial anisotropy field).⁷ $V^* \gg V$ means that several grains are exchange-coupled and switch together, $V^* \approx V$ means that grains switch almost independently, and $V^* < V$ means incoherent rotation. V^* is therefore the effective volume in magnetization reversal and controls the thermal stability⁸ and noise⁹ of magnetic recording media.

Nanostructured magnetic particles usually have grain sizes much smaller than the critical single-domain particle size and can be approximated by Stoner–Wohlfarth model.

V^* can be determined from magnetic viscosity (S) and irreversible susceptibility (χ_{irr}) measurements:¹⁰

$$V^* = k_B T \chi_{irr} / M_s S, \quad (1)$$

where T is the temperature and M_s is the saturation magnetization.

Generally in nanocomposite magnetic films, H_c increases with increasing grain volume V when the grains are isolated and exchange decoupled. However if V increases to such an extent that leads to the contact and strong exchange interaction between neighboring grains, an effective multidomain structure is formed and H_c will decrease rapidly with increasing V . When the grains have just started to contact each other and form a network-like structure, it is said that the percolation threshold is reached. Coercivity usually peaks near the percolation threshold.¹

In this article we report the structural and magnetic properties of Co:C nanocomposite films prepared by cosputtering and postannealing. The effects of annealing temperature, Co concentration, and film thickness on coercivity, saturation magnetization, physical grain volume, and activation volume are discussed. The effect of film thickness has become quite important because in extremely high density recording the film thickness has been reduced to about 15 nm,¹¹ which is comparable with the grain size.

II. EXPERIMENTAL METHODS

The Co:C films were cosputtered from a Co target and a graphite target onto water-cooled 7095 glass substrates. The base pressure was better than 3×10^{-7} Torr and Ar pressure was 10 mTorr. The Co:C layers are 5–100 nm thick with Co concentration varying from 50 to 80 at. %, and are protected by 20 nm C underlayer and overcoat. The as-deposited films were postannealed in vacuum at various temperatures

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FIG. 1. XRD patterns of 100 nm Co:C films with 60 at. % Co concentration annealed at various temperatures (T_A).

for two hours. The structural information was obtained from x-ray diffraction (XRD) and transmission electron microscopy (TEM) analyses. The hcp Co grain sizes of 100 nm films were estimated from the (100) peak width in XRD patterns by the Scherrer formula.¹² The magnetic measurements at room temperature were performed on an alternating gradient force magnetometer with the applied field parallel to the film plane. V^* was calculated from Eq. (1).

III. RESULTS AND DISCUSSION

Figure 1 shows the annealing-temperature (T_A) effect on the XRD patterns of 100 nm Co:C films with 60 at. % Co concentration. The as-deposited films are amorphous Co-C alloys as indicated in Fig. 1. Magnetic measurements show that they are nonmagnetic. This is in agreement with their amorphous structure. The Co_2C phase is formed as the majority phase at $T_A = 250^\circ\text{C}$. As T_A increases, Co_2C starts to decompose and isotropically oriented hcp Co crystallites are formed. TEM analysis confirms that Co_2C is the majority phase in films annealed at 250°C , and that hcp Co is the main crystalline phase in films annealed at 350°C . Figure 2 shows the high-resolution TEM pictures of a 100 nm film with 60 at. % Co concentration annealed at 350°C . It can be seen that the size of Co grains is in the order of 10 nm and some grains are in contact with each other. These results agree with other groups' work.^{4,13}

Saturation magnetization M_s , coercivity H_c , grain volume V , and activation volume V^* of 100 nm Co:C films with 60 at. % Co concentration are plotted in Fig. 3 as a function of annealing temperature T_A . M_s increases steadily as T_A increases from 250 to 350°C , then changes little with T_A . Because Co_2C is nonmagnetic, M_s directly indicates the amount of crystalline Co formed in the films. Thus it can be inferred from the M_s data that there is a small amount of crystalline Co in films annealed at 250°C . And as T_A increases, more crystalline Co is formed until T_A reaches

FIG. 2. High-resolution TEM picture of a 100 nm Co:C film with 60 at. % Co concentration annealed at 350°C .

400°C , when Co_2C has decomposed completely. Again this is consistent with the structural analysis discussed above.

Figure 3 also shows that with increasing T_A , V and V^* increase, while H_c first increases then decreases. The grain sizes estimated from the Scherrer formula range from 10 to 20 nm, which are much less than the film thickness. Spherical approximation is therefore used to estimate V . H_c reaches the maximum of about 800 Oe near $T_A = 300^\circ\text{C}$. The increase of V is caused by both the increase of the amount of crystalline Co and the growth of existing Co grains. But further increase of V brings the neighboring Co grains into contact. So near $T_A = 300^\circ\text{C}$ strong intergrain exchange coupling is developed and the percolation threshold is reached. This explains why the difference between V^* and V increases faster than V with increasing T_A when $T_A > 300^\circ\text{C}$. Now the T_A dependence of H_c can be understood. At low T_A , V and therefore H_c increase with increasing T_A . At high T_A , H_c decreases due to the formation of effective multidomain structure, as the percolation threshold is passed with increasing T_A .

FIG. 3. M_s , H_c , V , and V^* of 100 nm Co:C films with 60 at. % Co concentration annealed at various temperatures (T_A).

FIG. 4. M_s , H_c , V , and V^* of 100 nm Co:C films annealed at 400 °C with various Co concentrations.

In Fig. 4, M_s , H_c , V , and V^* of 100 nm Co:C films annealed at 400 °C are plotted as a function of Co concentration. As expected, M_s increases almost linearly with the increase of Co concentration. V and V^* also increase, but V^* increases much faster than V when Co concentration is higher than 60 at. %, suggesting that near this point strong intergrain exchange coupling occurs and the percolation threshold is reached. In Co:C films the 60 at. % Co concentration corresponds to 57 vol. % Co concentration, which is very close to the percolation threshold of ~ 55 vol. % found in Fe:SiO₂ system.¹ This result explains why at Co concentration of 60 at. % H_c reaches the maximum value of 790 Oe and from another aspect strengthens our reasoning of the V and V^* dependence of H_c discussed above.

Figure 5 shows the film thickness dependence of H_c in Co:C films with 60 at. % Co concentration annealed at various temperatures. The maximum H_c and therefore the percolation threshold occur in thinner film thickness when annealed at higher temperatures, indicating that in thinner films larger grains can be grown near the percolation threshold. This is understandable because the number of neighboring grains is reduced and percolation is formed only in a two-dimensional plane when the film thickness is comparable with the grain size. Similar results have been reported in Co:Ag granular systems.¹⁴ Moreover, the larger grains formed near the percolation threshold due to higher annealing temperature also lead to higher maximum H_c values. The highest H_c value of 1030 Oe was obtained in 20 nm films annealed at 450 °C. The films annealed at 550 °C is an exception, in which the maximum H_c is smaller than that of the films annealed at 450 °C. The reason is that the percolation threshold occurs at film thickness of about 5 nm. Even with large lateral grain size, the restriction in the film thickness still reduces the grain volume and consequently the value of maximum H_c .

IV. CONCLUSIONS

Nanostructured hcp Co particles with grain sizes ranging from 10 to 25 nm can be formed in Co:C composite films

FIG. 5. Film thickness dependence of H_c of Co:C films with 60 at. % Co concentration annealed at various temperatures.

upon appropriate annealing. Both higher annealing temperature and higher Co concentration result in larger grain size and stronger intergrain exchange coupling. High coercivities are developed in films with large grain and weak intergrain exchange coupling near the percolation threshold. By reducing the film thickness and increasing the annealing temperature, larger grain size and therefore higher coercivity can be obtained near the percolation threshold. This result is especially useful for the development of extremely high density recording media as the film thickness has been reduced to about 15 nm. The highest coercivity of 1030 Oe was obtained in 20 nm Co:C films with 60 at. % Co concentration annealed at 450 °C.

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