Intergranular interactions of low temperature atmosphere annealed Co/Pd magnetic multilayers

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We present evidence that low temperature atmosphere annealing of Co/Pd magnetic multilayers reduces both the dipolar and exchange interactions, but the dipolar interaction decreases very rapidly at the lowest temperatures. We also infer cobalt oxide formation at grain boundaries. This results in a reduction of the exchange between grains, reduces the length scale of ordering in the ac demagnetized state of the films. We demonstrate that this is the result of the decoupling the grains.
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INTRODUCTION

$(Co/Pd)_n$ magnetic multilayer thin films are candidates for media in high density magnetic recording technologies.[1–3] Such media rely on large surface anisotropies to achieve easy axes along the film normal, a condition necessary for both continuous and patterned perpendicular recording. We present a method of tailoring the properties of the magnetic multilayer film by thermal annealing.

In the annealing process, the lattice mobility of the atoms is enhanced, and the lattice will relax to a more energetically favorable state. However, should the annealing temperature be high enough and duration long enough, thermal agitation will result in considerable interdiffusion of the layers of the superlattice, reducing and eventually eliminating the advantages of depositing the multilayer structure.[4] In Yamane and Kobayashi have reported that during atmosphere annealing, oxygen seeps into the grain boundaries between the columnar Co/Pd multilayer grains and oxidizes with the cobalt so that its oxides, such as Co_3O_4, decouple the intergranular exchange, inhibiting domain wall propagation during magnetization reversal.[5]

To investigate these effects, we have annealed $(Co/Pd)_{30}$ multilayer thin films at 150, 200, and 250 °C for up to 2 h, in air and in this paper we propose to show that this modifies the microstructure of the film in such a way that intergranular interactions are reduced. This will be seen from the domain structure of ac demagnetized samples and the difference between the dc demagnetization (DCD) and initial remagnetization (IRM) curves in light of the measured microstructural properties of the annealed $(Co/Pd)_{30}$ multilayers. We will conclude that there are two regimes in the annealing process that affect intergranular interactions, one dominated by lattice relaxation and atomic migration and the other by oxidization and grain growth.

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SAMPLE PREPARATION

$(Co/Pd)_n$ magnetic multilayers were synthesized by sputtering at room temperature with a base pressure of less than 5×10^{-9} Torr on (100) Si with 5000 Å thermal oxide substrates, and a 20 nm layers of Ta to promote (111) growth of the fcc Co and Pd layers. The thickness of the Co and Pd layers was 4 and 11 Å, respectively, and each was deposited 30 times in succession. The multilayer thin films were then atmosphere annealed at 150, 200, and 250 °C for 5, 15, 35, 65, and 125 min.

RESULTS AND DISCUSSION

In order to investigate the effects of the microstructure on the magnetic properties of multilayer films, we initially characterize the structural properties themselves. Using x-ray diffractometery (XRD), we observe the rocking curve Pd (111) peaks of the samples annealed at 250 °C in Fig. 1. Figure 1(a) shows the raw data for the samples annealed at 250 °C, and in the compiled data of Fig. 1(b), we see that after an initial reduction in the linewidth of the spectrum, it increases. This is despite an initial reduction in the grain size, as seen in the scanning electron microscope (SEM) images. This indicates a considerable relaxation in the lattice strain at the onset of annealing, prior to the onset of significant diffusive effects that increase the grain size to 50 nm. For this reason we can separate the microstructural effects of annealing into an initial phase of diffusion and lattice relaxation and a later phase of grain growth and agglomeration.

The initial regime also accompanies a significant increase in the cobalt content at the surface. We observe this through X-ray photoelectron spectroscopy (XPS) studies of the Co $2p$ and the Pd $3d$ peaks in the $(Co/Pd)_{30}$ multilayers. The relative intensity of the cobalt peak to the palladium peak increases as we anneal the sample, indicating significant migration of cobalt atoms to the film surface. We will

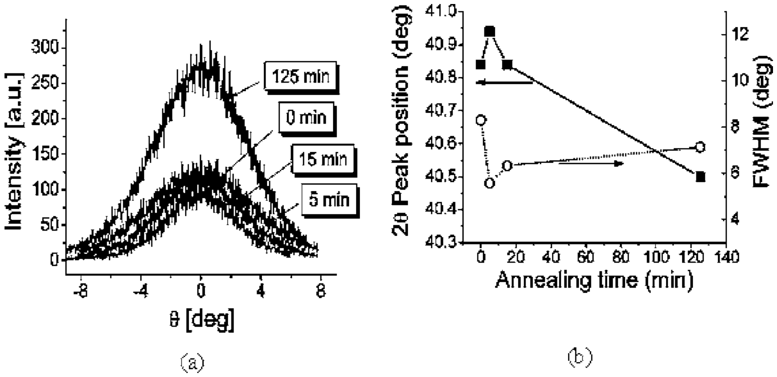

FIG. 1. (a) Rocking curves of (111) Pd peaks and (b) 2θ peak positions and full width at half maximum (FWHM) of rocking curves of samples annealed at 250 °C for 0, 5, 15, and 125 min.

attempt to show indirectly through magnetic microscopy that this migration is mainly cobalt oxides coating the grain boundaries.

We next turn to magnetic microscopy, both because it is our first indication that the grains are decoupling during the annealing process and because it is necessary to bolster our case that it is oxide in the grain boundaries that accounts for the decoupling. In this part, we will focus on the samples that were annealed at 250 °C. In Fig. 2 we reproduce magnetic force microscope (MFM) images after ac demagnetization. These show a change from long, uniform, and periodic stripe domains prior to annealing to small, irregular, and isolated domains after annealing. The long stripe domains are an indication of strong exchange coupling in the medium, and integration of the fast Fourier transform over all planar directions allows us to estimate the average width of the domains to be 250 nm. The increased dispersion in the periodicity of the demagnetized state, as seen in Fig. 3, indicates reduced exchange coupling across grain boundaries, indicating that the character of the grain boundaries has changed.

The remainder of the discussion will focus what we can infer from magnetometry. We have used the magneto-optical Kerr effect (MOKE) as well as vibrating sample magnetometry (VSM) to characterize these samples. In addition to the hysteresis loop measurements, summarized in Fig. 4, we have used magnetometry to acquire the DCD and IRM, as seen in Fig. 5. Examining the coercivity H_c in Fig. 4(a), we see the samples annealed at 250 °C see a reduction in H_c after 15 min of annealing. It is here where grain growth occurs, and therefore it is here that we associate with a change in the major effect of the annealing process. Comparison of the composition as measured by XPS to the coercivity in Fig. 4(b) indicates that the initial changes all scale with the migration of Co atoms. When the cobalt concentration surpasses 85% at the surface, the coercivity saturates. The change saturation magnetization M_s is just the opposite of the coercivity; very little decrease occurs in the early stages of the annealing process. This implies that oxidation only begins to have an effect on the magnetic atoms at this point, and a protective layer oxide must have been formed, as reported in Ref. 5. However, the major effects later in the annealing process are mainly the result of grain growth and oxidation.

The saturation magnetization has reduced, implying that intergranular coupling due to the demagnetization effect has likewise reduced, and so we turn to the DCD M_a and IRM M_r curves to help us measure the relative change in each, through the relevant difference in the two curves δM where $M_d(H)$ is the DCD remnant magnetization and $M_r(H)$ is the IRM initial remnant magnetization. In the Stoner-Wolfarth model, where there are no intergranular interactions, δM

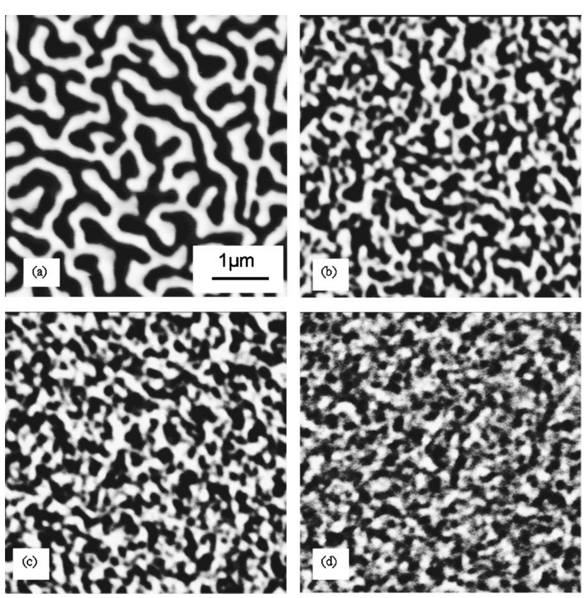

FIG. 2. MFM images of ac demagnetized states of sample 2 annealed at 250 °C for (a) 0 min, (b) 5 min, (c) 15 min, and (d) 125 min. 5×5 μm^2 scan. Switching from long range ordering to short range ordering as annealing time is observed.

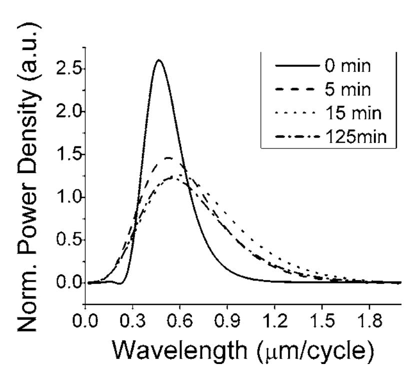

FIG. 3. Normalized two-dimensional (2d) power density spectrum of the MFM images for samples annealed at 250 °C for 0, 5, 15, and 125 min. The increased dispersion indicates the reduced exchange coupling across the grain boundaries.

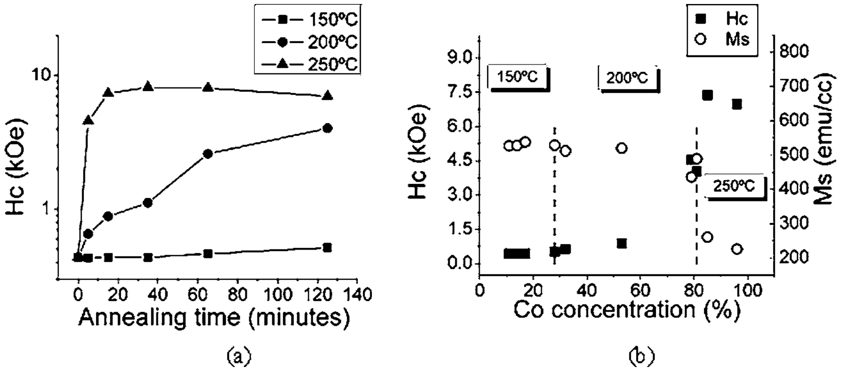

FIG. 4. (a) Coercivity (H_c) as a function of annealing time for Co/Pd multilayer thin film annealed at 150, 200, and 250 °C. (b) Coercivity and saturation magnetization as function of surface Co concentration evaluated from XPS measurements.

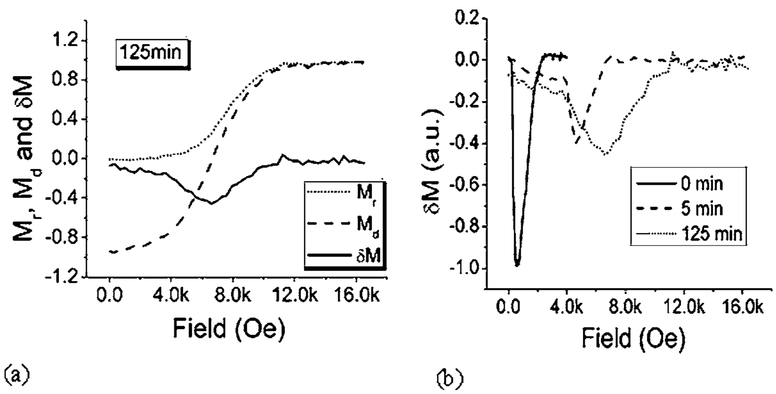

FIG. 5. DCD, IRM, and δM curves for Co/Pd multilayer thin film annealed at 250 °C for (a) 0 min and (b) 125 min.

vanishes. The effects of neighboring grains can be analyzed in terms of the deviation of δM from zero: positive deviations imply exchange interactions while negative deviations imply dipolar interactions.[6,7] We plot all three of these curves, normalized to M_s, in Fig. 5 for the unannealed sample and the samples annealed for 5 and 125 min at 250 °C. The shift of the δM curves indicates a change in the switching field distribution of the samples; however, changes in the shape of the curves, as analyzed in Ref. 6, show a continual decrease in the influence of exchange and the onset of the reduction in the magnetostatic interactions subsequent to the sample annealed for 5 min. These confirm the interpretations from MFM and hysteresis loop data.

CONCLUSION

We have investigated the intergranular interactions of low temperature annealed $(Co/Pd)_{30}$ multilayer thin films. The effects of annealing are separated into an early regime characterized by lattice relaxation and cobalt migration, and a later regime during which grain growth and oxide formation predominate. During the earlier period, slow cobalt migration to the grain boundaries reduces exchange between the grains, but the effect is striking even before measurable oxidation occurs. At the onset of the latter period, cobalt oxidation reduces the total moment of the sample very quickly, diminishing the magnetostatic energy, but this effect stops when there is enough oxide at the surface to protect the sample. The loss of the dipolar effects is mitigated by subsequent grain growth. These conclusions are supported by the difference between the DCD and IRM remnant magnetization curves. Low temperature atmosphere annealing is therefore a viable postsynthesis processing step to improve continuous perpendicular recording media.

ACKNOWLEDGMENTS

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