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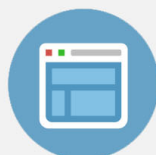
Effects of Ar-ion implantation and annealing on structural and magnetic properties of Co/Pd multilayers

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The contrasting effects of ion implantation and thermal annealing on structural and magnetic properties of Co/Pd multilayers have been studied. Ion implantation causes local damage to the multilayers, resulting in enhanced magnetization due to the polarization of the neighboring Pd. Thermal annealing generates massive interdiffusion across the interfaces into the formation of Co-Pd alloys with a lower magnetization. Effects on coercivity and remanence have also been studied.

Magnetic multilayers have been the subject of numerous studies[1,2] owing to their unusual properties and potential applications. Novel properties such as enhanced magnetization, large (sometimes perpendicular) magnetic anisotropy, tailored spin structures, and giant magnetotransport properties, have been uncovered. Among the multilayers, Co/Pd has been extensively studied in recent years,[3,4] stimulated by both fundamental interests and technological applications. For constituent layers with very small thicknesses, the perpendicular anisotropy together with substantial Kerr effect have led to the application of Co/Pd multilayers in magneto-optical recording media.[5]

While the novel properties of multilayers are intimately related to the high degree of structural coherency of the layered structure, a complete understanding of the relationship between interfacial characteristics and physical properties remains lacking. In this work we have systematically altered the interfacial characteristics of Co/Pd multilayers using ion implantation and thermal annealing, and studied the resultant structural and magnetic properties. Our studies show that these two methods give rise to very different effects on the multilayer structure.

The Co/Pd multilayer samples have been deposited onto glass substrates using two electron-beam-deposition sources in a vacuum of 1×10^{-8} mbar. The samples are denoted as $[Co(x \text{ Å})/Pd(y \text{ Å})]_n$, where x and y are the Co and Pd layer thicknesses, respectively, and n is the number of repeats of the Co/Pd bilayers. One set of samples with $[Co(10 \text{ Å})/Pd(59 \text{ Å})]_{18}$ has been subjected to 230 keV of Ar^+ implantation at room temperature at a low current density of 50 nA/cm^2 to avoid sample heating. Low fluences of 1×10^{13}–5×10^{14} ions/cm^2, which are small compared with 1 monolayer of ions (1×10^{15} ions/cm^2), have been used. Another set of samples of $[Co(14 \text{ Å})/Pd(44 \text{ Å})]_{18}$ have been thermally annealed for 30 min in a helium atmosphere at various temperatures up to 700 K. Structural characteristics of all the samples have been made by a θ-2θ x-ray diffractometer with $CuK\alpha$ radiation. Magnetic properties have been measured using a vibrating sample magnetometer (VSM) with the magnetic field parallel to the film plane.

Figure 1 shows the x-ray-diffraction data of the as-prepared and the ion-implanted $[Co(10 \text{ Å})/Pd(59 \text{ Å})]_{18}$ samples. The as-prepared sample [Fig. 1(a)] shows several

satellite peaks near the Pd(111) peak. Because Co layers are thin and have a much lower scattering factor, no substantial diffraction peaks near the Co(111) peak are revealed nor expected. It is well known that the intensity of the satellite peaks is dictated by the layer profile and the repeatability of the bilayers. The structural coherence of the multilayers is manifested by the width of the diffraction peaks. From the width of the main diffraction peak, beyond the instrumental width, the structural coherence length L has been determined to be about 270 Å, which is about four bilayers. These results show that the as-prepared sample has a well-defined layer structure with [111] crystalline orientation.

Figures 1(b)–1(e) show the diffraction data for ion-implanted samples of $[Co(10 \text{ Å})/Pd(59 \text{ Å})]_{18}$ with Ar fluence varying from 5×10^{13} to 7.5×10^{14} ions/cm^2. Several features are noted for samples within this fluence range. For increasing fluence, the satellite peaks decrease progressively in intensity but remain observable in all cases. More important, the width of the main diffraction stays essentially the same as that of the as-prepared sample. These results indicate that layer structure and the structural coherence length L remain intact, but the layer profile becomes more diffuse as a result of ion implantation.

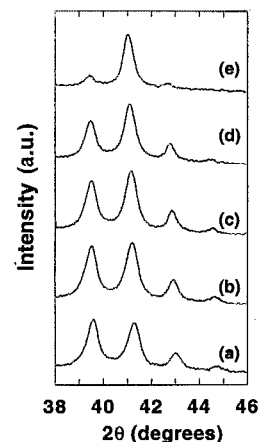A contrasting situation occurs when the Co/Pd multilay-

FIG. 1. X-ray spectra of (a) as-prepared sample and the implanted samples: (b) 5×10^{13}, (c) 1×10^{14}, (d) 5×10^{14}, and (e) 7.5×10^{14} ions/cm^2.
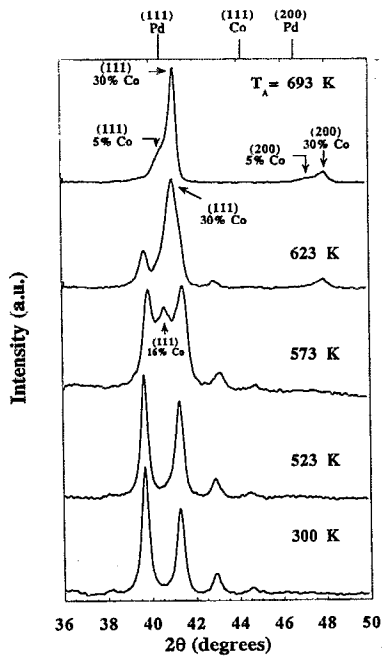
FIG. 2. X-ray spectra of samples annealed at different temperatures T_A. The peak positions corresponding to bulk Co and Pd are shown at the top. Alloys with different compositions have also been indexed.

FIG. 3. Saturation magnetization in unit of emu/cm^3 of Co at 300 K (dotted curves) and 73 K (solid curves) as functions of fluences (left-hand side) and annealing temperatures (right-hand side).

ers have been thermally annealed at various annealing temperatures T_A. The x-ray-diffraction data of the as-prepared (T_A=300 K) and representative data of the annealed samples (T_A up to 693 K) of [Co(14 Å)/Pd(44 Å)]$_{18}$ are shown in Fig. 2. Discernible change of the diffraction are observed for $T_A \geqslant 523$ K; most notably a diminution of the satellite peak intensity and the appearance of additional diffraction peaks. It is useful to recall that Co and Pd form fcc Co-Pd alloys over the entire composition range. The lattice parameter of the alloy decreases monotonically from that of pure Co to that of pure Pd. At the top of Fig. 2 the locations of the diffraction peaks of Co(111) and Pd(111) are shown, and the (111) peak of a Co-Pd alloy falls between those of Co(111) and Pd(111) according to the composition. At T_A=573 K, in addition to the satellite peaks, a new peak, corresponding to Co$_{16}$Pd$_{84}$, appears. At T_A=623 K both the (111) and (200) peaks of Co$_{30}$Pd$_{70}$, in addition to the remanence of the satellite peaks, are observed. Finally, at T_A=693 K, only (111) and (200) peaks corresponding to predominantly Co$_{30}$Pd$_{70}$ and a small portion of Co$_5$Pd$_{95}$ are observed. There are no satellite peaks remaining, and the layer structure has been completely destroyed. It may be noted that, if the Co and Pd atoms in [Co(14 Å)/Pd(44 Å)]$_{18}$ were mixed completely and uniformly, one would have obtained an alloy of Co$_{30}$Pd$_{70}$, which is very similar to the alloy composition found.

From the above results, it is clear that both ion implantation and thermal annealing can cause deterioration of the multilayers; however, the processes with which the layer structure and the interfaces are compromised are very different in the two cases. At low fluence, ion implantation causes local damages of the layer structure, while most of the layer structure and the structural coherence of the multilayer re-
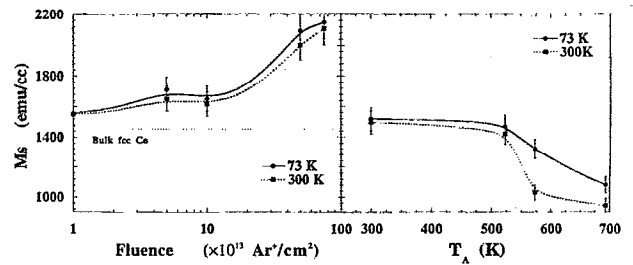
main intact. On the other hand, thermal annealing is far more disruptive. It promotes interdiffusion of all the constituent atoms at the interfaces, very efficiently destroying the multilayers even at moderate annealing temperatures. Furthermore, due to the interdiffusion, the multilayer rapidly converges to the terminal and homogeneous alloy composition.

These conclusions made from structural studies are corroborated by the magnetic properties. It is useful first to recall the magnetic properties of Co$_x$Pd$_{100-x}$ alloys of which the magnetic ordering temperature T_C increases rapidly and monotonically with Co content. The values of T_C are less than 300 K for the alloys with low Co content (x<10).[6] The magnetic moment per atom of Co-Pd alloys also rises monotonically with Co content. However, because of the strong polarization effect on the Pd atoms by the nearby Co atoms (also known as the giant moments), these polarized Pd atoms contribute significantly to the magnetization.

The saturation magnetization M_s of the Co/Pd multilayers after ion implantation and thermal annealing are shown in Fig. 3. First of all, the as-prepared samples have a higher M_s than that of bulk fcc Co, owing to the well-known polarization effect of Pd. The enhanced magnetization has also been observed in other Co/Pd and Fe/Pd (Ref. 7) multilayers. Most remarkably, after ion implantation, there is further enhancement of M_s, and M_s has a weak temperature dependence; the values at 77 and 300 K are similar. This indicates that, as the interfaces are systematically disrupted by ion implantation, more Pd atoms are exposed and polarized by the displaced Co, hence a larger M_s. Furthermore, most of the magnetic species remain strongly coupled to share a high value of T_C. Quite the contrary, the thermally annealed samples show a decrease in M_s and a stronger temperature dependence in M_s. Both of these observations, which are consistent with another group's results,[8] are the results of the formation of Co-Pd alloys with relatively low Co contents, hence lower values of T_C.

In Fig. 4, the magnetic coercivity H_c for the implanted [Fig. 4(a)] and the annealed [Fig. 4(b)] samples are shown. We discuss the annealed samples first. Changes in H_c occur when the formation of the Co-Pd alloy appears at $T_A \geqslant 523$ K. Higher values of T_A result in large values of H_c, and H_c shows similar behaviors at the three temperatures (77, 150, and 300 K) measured. For the ion-implanted samples, H_c for the ion-implanted samples with higher fluences ($\geqslant 5 \times 10^{14}$
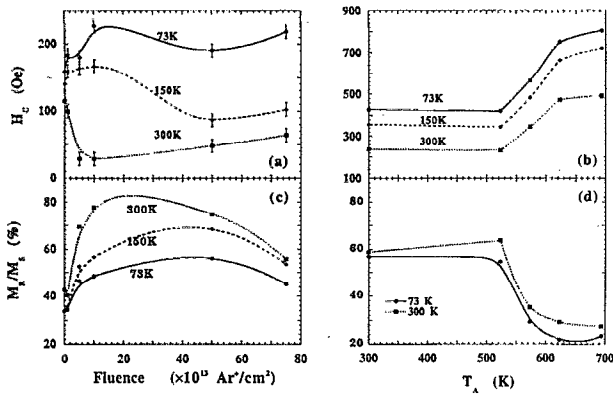
FIG. 4. In-plane coercivity H_c and squareness M_r/M_s at different temperatures as functions of fluences (left-hand side) and annealing temperatures (right-hand side).

presence of Co-Pd alloy phases. In the annealed samples, the predominant evolution of the multilayer is toward the formation of Co-Pd alloys, which generally give rise to lower squareness.

In conclusion, we have observed via structural and magnetic properties the very different effects on the Co-Pd multilayer due to ion implantation and thermal annealing. Ion implantation causes local damage to the interfaces while maintaining the layer structure, whereas thermal annealing promotes massive interdiffusion toward the formation of Co-Pd alloys. Consequently, contrasting behaviors in saturation magnetization and other magnetic properties have been observed.

ions/cm^2) has similar temperature dependencies as the annealed samples. The results for the low fluences are, however, more complex. In particular, the results of the implanted samples with fluences less than 1×10^{14} ions/cm^2 suggest the possibility of a phase(s) with low ordering temperature (<300 K). The precise nature of these possible phases, albeit of low sample content, has not been ascertained.

Finally, we turn to remanent magnetization M_r expressed as the squareness M_r/M_s shown in Figs. 4(c) and 4(d). After ion implantation with low fluence, the squareness becomes larger, suggesting an increase in the in-plane anisotropy. At high fluence, the squareness decreases due to the

[1] See, for example, Physics, Fabrication and Applications of Multilayered structures, edited by P. Dhez and C. Weisbuch (Plenum, New York, 1988).
[2] L. M. Faclicov et al., J. Mater. Res. 5, 1299 (1990).
[3] P. F. Carcia, A. D. Meinhaldt, and A. Suna, Appl. Phys. Lett. 47, 178 (1985).
[4] B. N. Engel, C. D. England, R. A. Van Leeuwen, M. H. Wiedmann, and C. M. Falco, J. Appl. Phys. 70, 5873 (1991).
[5] M. J. A. M. Greidanus and W. B. Zeper, MRS Bull. 15, 31 (1990).
[6] R. M. Bozorth, P. A. Wolfe, D. D. Davis, V. B. Compton, and J. H. Wernick, Phys. Rev. 122, 1157 (1961).
[7] F. J. A. den Broeder, H. C. Donkersloot, H. J. G. Draaisma, and W. J. M. de Jonge, J. Appl. Phys. 61, 4317 (1987); J. R. Childress, A. Schuhl, J.-M. George, P. Durand, P. Galtier, V. Cros, K. Ounadjela, R. Kergoat, and A. Fert, in Magnetism and Structure in Systems of Reduced Dimension (Plenum, New York, 1993).
[8] F. J. A. den Broeder et al., J. Phys. (Paris) Colloq. 8, C-1663 (1988).