Magnetization and anisotropy of Co/Pd multilayer thin films

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Multilayered Co/Pd thin films were prepared by sequential electron-beam evaporation of Co and Pd onto Si substrates at room temperature. The thicknesses of the Co sublayer and of the Pd sublayer were varied between 2.0–10.3 and 4.5–22.3 Å, respectively. As the Pd sublayer thickness was varied at constant Co thickness, broad maxima in the saturation magnetization $M_s$ and intrinsic perpendicular anisotropy energy $K_u$ were observed at a Pd thickness of about 10 Å. At this maximum, $M_s$ per Co volume is larger than the saturation magnetization of bulk Co. This is believed to be caused by the polarization of the Pd atoms within about 10 Å of the Co layer. $K_u$ and $M_s$ per Co volume both decrease with increasing Co layer thickness.

I. INTRODUCTION

Magneto-optic recording systems, based on rare-earth transition-metal (RE-TM) alloy recording media, have recently become commercially available. Although the RE-TM alloys have many desirable and easily adjusted properties, they have two primary weaknesses. First, the signal level that can be obtained from these materials, proportional to $R\delta$, where $R$ is the reflectivity and $\delta$ is the Kerr rotation, is relatively small, on the order of 0.25. $R\delta$ decreases with decreasing wavelength. Although adequate for existing products, this limits future improvements. Second, RE-TM materials are highly susceptible to corrosion, which can impair the magnetic and optical properties or create error-causing defects. RE-TM alloys have been extensively studied and, after great initial strides, progress in remedying these defects has waned.

Over the past several years techniques to produce novel materials with customized properties have flourished. Chief among these is the development of compositionally modulated structures (CMS). In a CMS the composition is varied in a controlled manner on an atomic scale, allowing materials, crystal structures, and properties not found using bulk techniques. One particular class of CMS involves alternate layers of a magnetic and a nonmagnetic material. Recently, Garcia, Meinhaldt, and Suna reported that Co/Pd CMS films had perpendicular magnetic anisotropy, a property not found in Co/Pd alloys. Shin and Palumbo report that CMS Co/Pd shows a significant Kerr rotation (optically thick films consisting of 2-Å-thick Co and 9-Å-thick Pd layers had $2\delta_e = 0.24^\circ$ when the ellipticity was removed). Compositonally modulated structures seem a promising approach to achieving stable, high-performance magneto-optical recording materials.

II. EXPERIMENTAL METHODS

Co/Pd multilayer thin films were prepared by e-beam evaporation of Co and Pd onto Si substrates in a vacuum system maintained at about $2 \times 10^{-6}$ Torr during the deposition. Substrates were at room temperature. The compositionally modulated structure was obtained by alternately exposing the substrate to the two sources via a rotating substrate mount. The two e-beam sources were physically separated by stainless-steel shields to prevent cross-contamination of their evaporated fluxes. The sublayer thickness of each constituent in one repeat distance was varied by changing the deposition rate of the corresponding source. The deposition rate of each source was monitored by a quartz-crystal sensor. Typical deposition rates of Co and Pd were 0.9–2.8 and 2–10 Å/s, respectively. The thicknesses of the Co sublayer and of the Pd sublayer were varied between 2.0–10.3 and 4.5–22.3 Å, respectively. Sublayer thicknesses are accurate to within 10%. The number of bilayers was varied to maintain a total film thickness of approximately 1000 Å.

X-ray diffraction demonstrated that the films were polycrystalline fcc with a (111) orientation and a lattice constant intermediate between Co and Pd. Satellite lines indicative of a superlattice structure were observed; extensive analysis of the quality of the interfaces was not performed.

Magnetization was measured using a vibrating sample magnetometer (VSM) calibrated against a Ni standard. The maximum field used was at least 13 kOe. The anisotropy was measured using a torque magnetometer and the data analyzed according to the method of Miyajima and Sato. Torque measurements were done at a field of 19 kOe. For both magnetization and anisotropy measurements, the small signal from the sample holder and uncoated substrate was subtracted out. Kerr rotation measurements were also made on these samples; the results are reported elsewhere.

III. RESULTS AND DISCUSSION

A minimum requirement for a magneto-optic storage medium is perpendicular anisotropy and a square hysteresis loop. Squareness is defined by the ratio of the remnant magnetization to the saturation magnetization, $M_r/M_s$. Figure 1 displays the measured squareness of the VSM-determined hysteresis loop as a function of Co and Pd sublayer thickness when the applied magnetic field is perpendicular to the substrate. Clearly, square loops are obtained when the Co layer thickness is approximately 2 Å and the Pd layer thickness is greater than 5 Å. Under these conditions the squareness is close to 1. Squareness decreases with increasing Co thickness and decreasing Pd thickness.

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At fixed Co thickness, $M_s$ decreases with increasing Pd thickness, except there is an indication of a slight maximum in $M_s$ near a Pd thickness of 10 Å. When the Co thickness is 2 Å, a pronounced maximum in $M_s$ is observed when the magnetization per volume of Co is plotted as in Fig. 2. The maximum $M_s$/Co at a Pd thickness of 9 Å and a Co thickness of 2 Å exceeds 1422 emu/cm$^3$, the value for crystalline Co at room temperature. Our films are polycrystalline and show voids when observed by transmission electron microscopy; the pure Co films have reduced magnetization relative to the bulk, which accounts for the observation that $M_s$/Co does not approach the value for bulk Co as the Pd thickness approaches zero. In addition, $M_s$/Co is seen to decrease slightly with increasing Co thickness.

Figure 3 shows both the saturation magnetization per Co volume and the saturation Kerr rotation as a function of Pd thickness for a constant 2-Å Co thickness. The dilution effect has been removed by dividing the Kerr rotation by the Co sublayer thickness to bilayer thickness ratio. The maximum in magnetization is clearly present while the Kerr rotation increases slightly with increased Pd thickness. It is reasonable to assume that the Kerr rotation is due to the Co, and therefore the magnetic and optical properties of the Co layers are not changing significantly. This is consistent with the fact that Co is a saturated ferromagnet. Consequently the enhanced magnetization must be due to polarization of the Pd.

Enhanced magnetization caused by the polarization of Pd has been reported in CoPd alloys, and in Co/Pd multilayer films. Broeder et al. assume the layer of Pd adjacent to the Co (2.15 Å) is polarized and find the magnetization per Pd atom to be 0.6 $\mu_B$ in agreement with Bozworth. Garcia assumes the Pd magnetization is

$$M_{Pd}(x) = \left[ A \cosh(x/l) / \cosh(t_{Pd}/2l) \right],$$

where $x$ is the distance from the center of the Pd layer, $A$ is the magnetization of the Pd adjacent to the Co layer, $l$ is an adjustable interaction length, and $t_{Pd}$ is the thickness of the Pd layer. The best fit is obtained with $A = 856$ emu/cm$^3$ and $l = 5.3$ Å. In either case it is predicted that the magnetization per volume of Co should increase with increasing Pd thickness until the interaction length is exceeded, after which it should be constant. The data reported by Broeder and by Garcia are consistent with their interpretation. Our data also show $M_s$/Co increasing with increasing Pd thickness, but only up to a Pd thickness of 9 Å. The decrease we observe in $M_s$/Co for Pd thicknesses above 9 Å could be caused by an antiferromagnetic interaction with the more distant Pd atoms, or more likely, a change in the structure of the Pd layers or the lowering of the Curie temperature with increased Pd content.

The uniaxial anisotropy constant $K_u$ per total volume varies with Co and Pd thickness in a manner qualitatively similar to the magnetization per Co volume, decreasing slowly with increasing Co and displaying a pronounced maximum at a Pd thickness of 9 Å. The decrease with increasing Co thickness is greatly accentuated when $K_u$ per Co volume is examined. For a fixed Co thickness of 2 Å, these two measures of $K_u$ are plotted as a function of Pd thickness in Fig. 4. As mentioned above, $K_u$ per total volume displays
a clear maximum at the same Pd thickness as does the magnetization. $K_a$ per Co volume also drops rapidly for Pd thicknesses below 9 Å, but decreases only slightly for Pd thicknesses above 9 Å. Therefore, for $t_{pd} > 9$ Å the anisotropy energy per Co/Pd interface area is constant, indicating the interface is the source of the anisotropy.

For a Pd thickness of 9 Å and above, these values of $K_a$ per cobalt volume agree with those reported by Draaisma and co-workers. Following Eq. (1) of that reference, we write $K_a = 2K_r/t_{Co} + K_r$, where $K_a$ is the anisotropy energy per Co volume, $K_r$ is the anisotropy originating from the surface per unit area, $t_{Co}$ is the Co thickness, and $K_r$ is the volume anisotropy including demagnetization energy, magnetocrystalline anisotropy, and magnetoelastic energy. By plotting $K_a$ vs $1/t_{Co}$, the values of the various terms can be obtained. With only three Co thicknesses for $t_{pd} = 9$ Å and two for $t_{pd} = 4.5$ Å, our data do not include sufficient Co thicknesses for an accurate determination of the parameters, but we do obtain a linear relationship. Unlike Draaisma and co-workers or Carcia and co-workers, our values depend on Pd thickness and are shown in Table I.

### IV. CONCLUSION

The use of a compositionally modulated structure induces perpendicular anisotropy in Co/Pd with a well-defined maximum in magnetization and anisotropy when the Co layer thickness is 2 Å and the Pd layer thickness is 9 Å. The results are consistent with the Co-Pd interface being the source of the anisotropy. For certain values of Pd thickness, the magnetization per Co volume exceeds that of bulk Co; this is due to polarization of the Pd atoms. Unlike previous investigators, we observe a decrease in $M_s/Co$ as the Pd thickness is increased above 9 Å. This is not consistent with either the polarization of just the Pd atoms adjacent to the Co layer, or with a $\cosh(x)$ dependence of the Pd polarization.