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Magnetocrystalline Anisotropy of Magnetic Grains in $\text{Co}_{80}\text{Pt}_{20}$:Oxide Thin Films Probed by X-ray Magnetic Circular Dichroism

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Abstract

Using angle-dependent X-ray magnetic circular dichroism we have measured magnetic hysteresis loops at the $\text{Co}L_{2,3}$ edges of oxide-doped $\text{Co}_{80}\text{Pt}_{20}$ thin films. The magnetocrystalline anisotropy energy (MAE) of the Co atoms, which is the main source of the magnetocrystalline anisotropy of the CoPt magnetic grains, has been determined directly from these element-specific hysteresis loops. When the oxide volume fraction (OVF) is increased from 16.6% to 20.7%, the Co MAE has been found to decrease from 0.117 meV/atom to 0.076 meV/atom. While a larger OVF helps to achieve a smaller grain size, it reduces the magnetocrystalline anisotropy as demonstrated unambiguously from the direct Co MAE measurements. Our results suggest that those $\text{Co}_{80}\text{Pt}_{20}$:oxide films with OVF between 19.1% and 20.7% are suitable candidates for high-density magnetic recording.

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I. Introduction

In order to increase areal densities in hard disk drives and to achieve narrower bit boundaries, it is essential to reduce the magnetic grain size as well as the exchange and/or magnetostatic interaction between the grains^{1, 2}. The oxide based grain boundary is effective for this purpose, e.g., oxygen³ or SiO₂⁴ has been composited into CoPtCr to reduce grain size and inter-granular exchange coupling by oxide materials that easily precipitate at the grain boundary.

Girt *et al.*⁵ succeeded in isolating the magnetic grains by co-sputtering Co and Pt with nonmagnetic oxide material which serves as a barrier to decouple neighboring grains. With increasing oxide volume fraction (OVF) the magnetic grain size becomes smaller, while at the same time the magnetocrystalline anisotropy of the grains, K_{1g} , was found to be significantly reduced⁵. This result is critical as it suggests that there is a limit for the areal density capability caused by the dilemma between the simultaneous requirements for small grain size and large K_{1g} . The value of K_{1g} is usually *calculated* from the magnetocrystalline anisotropy of the media, K_1 , by taking account of the OVF^{5, 6}. However, a direct observation and accurate experimental determination on the magnetic properties and anisotropies of the magnetic grains is lacking and necessary.

Element-specific X-ray magnetic circular dichroism (XMCD) offers a unique tool for this purpose, as it is capable of measuring the magnetization loops of the individual magnetic element in the magnetic grains, excluding those contributions from the oxide material. In this paper, we measured the angle-dependent XMCD hysteresis loops at the Co $L_{2,3}$ edges for a series of Co₈₀Pt₂₀:oxide thin films with variable OVF of ~16.6% to 20.7%. In $3d$ - $5d$ transition metal alloys, such as Co-Pt, the net magnetization mainly arises from the Co $3d$ magnetic moment, while the Pt $5d$ states carry a small component induced by the $3d$ - $5d$ hybridization⁷. This means that despite the important role of the hybridization in giving rise to the giant magnetocrystalline anisotropy of the Co-Pt systems⁸, the magnetocrystalline anisotropy energy (MAE) of the Co atoms is still the main source of the K_{1g} ⁹, which can be determined directly from the element-specific hysteresis loops. In this work, we

have found that when the OVF increases from 16.6% to 20.7%, the Co MAE decreases from 0.117 meV/atom to 0.076 meV/atom. In combining with the magnetic measurements from vibrating sample magnetometry (VSM), this work has further suggested that an OVF range of 19.1% - 20.7% is suitable for the magnetic recording.

II. Sample Preparation

We prepared a series of samples, glass/Ta (5 nm)/Ru (13 nm)/Co₈₀Pt₂₀ + oxide (total of 13 nm)/C (7 nm), grown by the same method as described in Ref. 5. The samples were deposited at room temperature using dc and rf magnetron sputtering in a Unaxis M12 sputter tool with base vacuum below 1×10^{-6} Pa. The magnetic layer of each sample was fixed at the same thickness, formed by co-sputtering Co, Pt, and oxide targets. The CoPt grains were grown on top of Ru grains, with oxide material segregation to the grain boundaries. The oxide co-sputtering power was varied from 5 W to 30 W, which controls the OVF and the grain size, as described below.

III. Results and Discussion

A. TEM measurement of OVF

Plane-view transmission electron microscopy (TEM) was performed to investigate the OVF outcome and corresponding grain sizes. As shown in Fig. 1, the magnetic grains are well isolated by oxide material segregated into the grain boundary. The grain size distribution of this granular layer has been fitted with a log-normal distribution and is shown in the inset, and the mean grain size D is estimated to be 7.8 nm with a standard deviation of ± 1.6 nm.

From the TEM analysis, we found that the OVF and grain size dependences are almost linear with the oxide power. As the power increases from 5 W to 30 W, the OVF in the magnetic layer increases from 16.6% to 20.7%, and the grain size reduces from 10.0 nm to 7.7 nm, namely, the grain sizes D are 10.0 nm, 9.5 nm, 8.6 nm, and 7.7 nm, respectively, for the OVF of \sim 16.6%, 17.5%, 19.1%, and 20.7%.

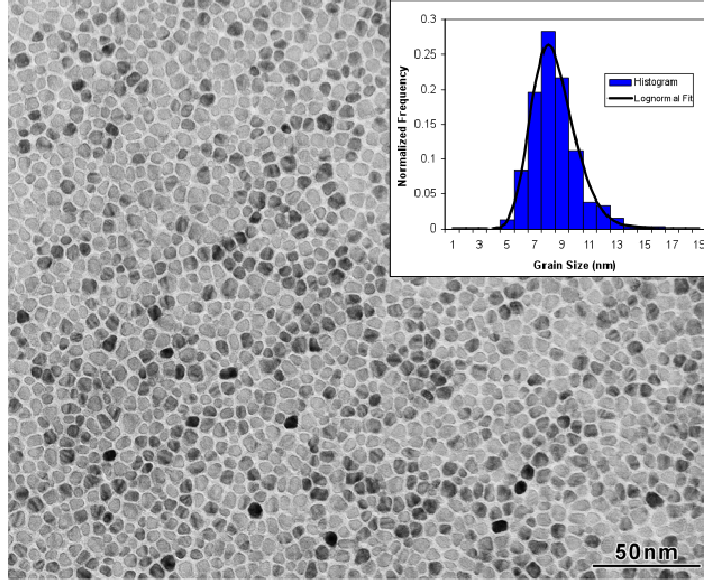


FIG. 1: (Color online) Plane-view TEM image of the granular layer for oxide power of 28 W. Inset: From the grain size distribution of the granular layer the meangrain size D is estimated to be 7.8 nm.

B. VSM results as a function of OVF

Figure 2(a) shows a typical hysteresis loop of the sample with OVF $\sim 16.6\%$ measured by VSM, with the magnetic field applied along the film normal. The coercivity H_C , nucleation field H_N and remanence magnetization M_r are marked on the loop. H_N is defined as the field of the intercept between the saturation magnetization level and the tangent at H_C ¹⁰, so that this property describes how well the medium is able to resist the erasing field from the return pole during the writing process. One more important parameter, remanence squareness S , is defined as the ratio of the remanence magnetization M_r against the saturation magnetization M_S ¹¹, i.e., $S = M_r / M_S$.

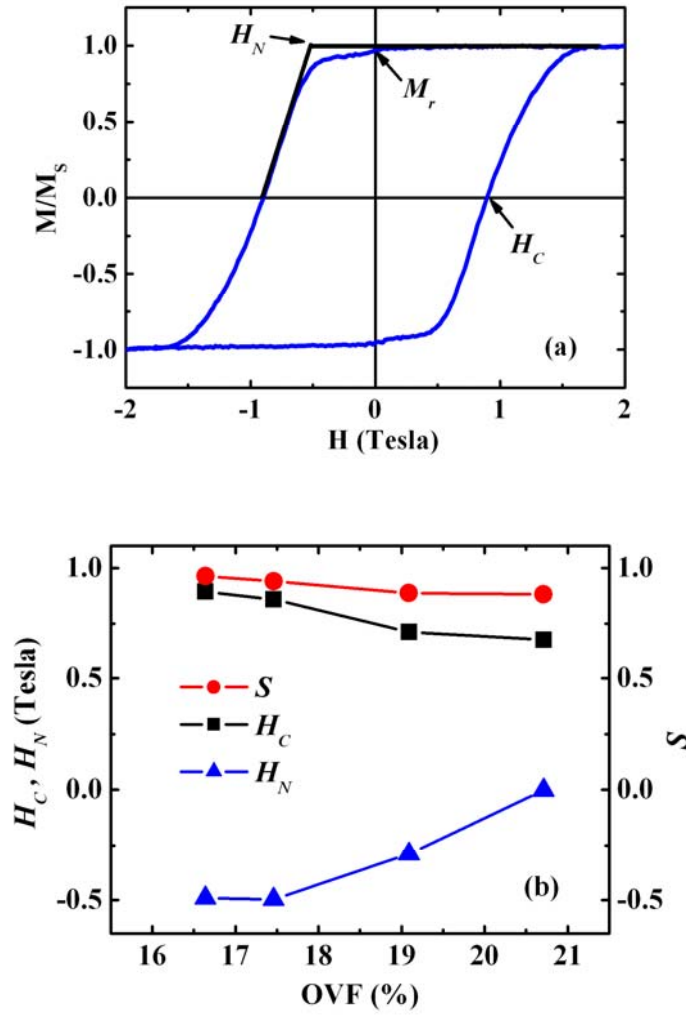


FIG. 2: (Color online) (a) Hysteresis loop measured by VSM when OVF $\sim 16.6\%$, and definition of coercivity H_c , nucleation field H_N and remanence magnetization M_r ; (b) H_c , H_N , and remanence squareness $S = M_r/M_s$, along the normal direction as a function of OVF.

The trends of H_c , H_N , and S are shown in Fig. 2(b) as functions of OVF. For increasing OVF the coercivity H_c decreases and the nucleation field H_N increases, which suggests that the recording-layer grains are magnetically exchange decoupled and broken up due to enhanced segregation between the media grains.¹² It is commonly acknowledged

that a large negative H_{Ni} is essential for the stability of the recorded bit¹⁰, and as one can see in Fig. 2(b), here H_{Ni} is below zero when the OVF is lower than 20.7%, reaching -0.29 T when OVF \sim 19.1%. The value of S reaches almost 1.0, indicating a high potential of thermal stability of the read-back signal even at low recording density¹³. The decreased S for increasing OVF indicates a reduced intergranular exchange coupling, which is expected to lead to the formation of smaller domains and in this case the magnetic grains may be reversed individually.¹⁴

C. MAE as a function of OVF

XMCD measurements were performed on bending magnet beamline 6.3.1 at the Advanced Light Source, Berkeley, using circularly polarized X-rays with a degree of circular polarization of \sim 60%. Using the magnetic field reversal method with a field of \pm 1.8 T applied parallel to the direction of the incident X-rays, XMCD¹⁵ spectra were recorded at the Co $L_{2,3}$ edges^{16, 17, 18} and the spin and orbital moments were evaluated by sum rule analysis¹⁹. Figure 3 shows the normalized XMCD spectra measured at the Co $L_{2,3}$ edges at angles $\gamma = 0^\circ$ (normal) and $\gamma = 60^\circ$ (grazing) with respect to the surface normal of the sample with 19.1% OVF.

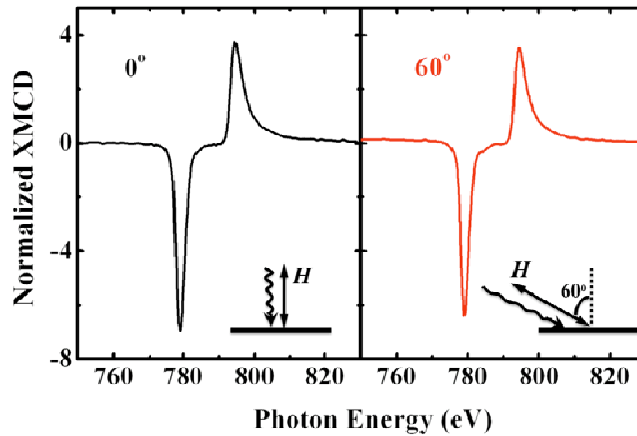


FIG. 3: (Color online) Normalized XMCD spectra at the Co L_3 (\sim 778 eV) and L_2 (\sim 793 eV) edges for the CoPt thin film with 19.1% OVF. Spectra are shown for normal (black) and grazing (red) X-ray incidence relative to the

surface. In both cases the ± 1.8 T magnetic field was along the X-ray incidence direction.

We measured element-specific magnetic hysteresis loops by recording the peak height of the $\text{Co}L_3$ XMCD signal at ~ 778 eV divided by the peak height of the $\text{Co}L_2$ XMCD signal at ~ 793 eV as a function of the applied magnetic field, for $\gamma = 0^\circ$ (circles) and 60° (squares), as shown in Fig. 4. The curves show a distinct hysteresis, revealing that the CoPt grains retain grain-grain ferromagnetic alignment at room temperature and all samples exhibit an out-of-plane easy axis.

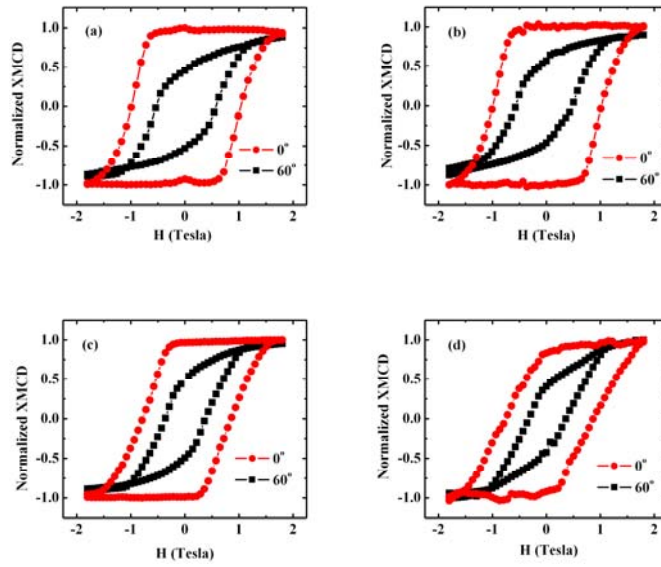


FIG. 4: (Color online) XMCD magnetic hysteresis loop for CoPt films with OVF = (a) 16.6%, (b) 17.5%, (c) 19.1%, and (d) 20.7%, measured at the $\text{Co}L_{2,3}$ edges as a function of the applied magnetic field at angles $\gamma = 0^\circ$ (red circles) and 60° (black squares).

Figure 5 shows the coercivity of the samples determined from the XMCD hysteresis loops, $H_{C,\text{XMCD}}$, at $\gamma = 0^\circ$ (red closed circles) and 60° (black closed squares). The values and trend are both similar to those measured by VSM, indicating

that the main contribution of the total magnetic moments is due to Co. The theoretical curves of $H_{C,XMCD}$ for $\gamma = 60^\circ$ using the domain wall motion (DWM) model (blue open triangles) and the Stoner-Wohlfarth (SW) rotation model (green open triangles) are plotted for comparison²⁰. We see that the experimental data of $H_{C,XMCD}$ at $\gamma = 60^\circ$ almost fully agrees with the calculation using the SW rotation model. However, when the OVF is lower than 19.1%, the magnetization reversal process is slightly towards DWM due to insufficient isolation of the CoPt grains. When the amount of oxide is insufficient to surround the CoPt grains, the oxide may exist as discontinuous sheets or clusters, which behaves as pinning sites and leads to a large coercivity during DWM.²¹ When OVF is increased to and larger than 19.1%, more oxide resides around the grain boundaries and forms a quasi-continuous network, whose size might be too large to effectively pin the domain wall, resulting in a smaller $H_{C,XMCD}$ value.²²

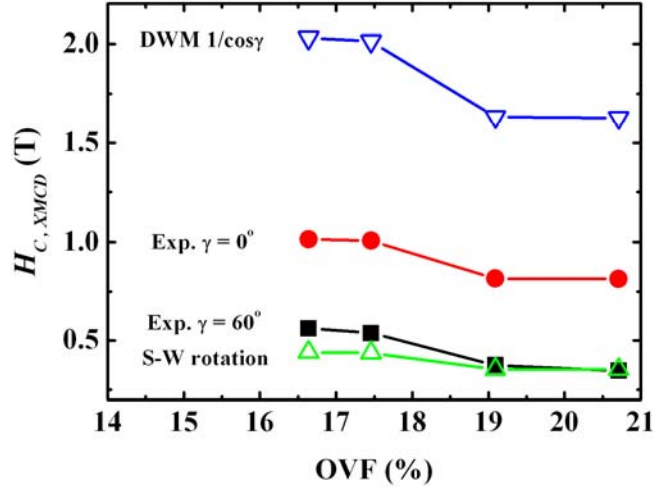


FIG. 5: (Color online) Coercivity measured by XMCD hysteresis loop ($H_{C,XMCD}$) for CoPt films with variable OVF at $\gamma = 0^\circ$ (red closed circles) and 60° (black closed squares). Also plotted are theoretical curves for $H_{C,XMCD}$ at $\gamma = 60^\circ$ using the DWM model (blue open triangles) and SW rotation model (green open triangles).

As expected, the reversal process of these samples tends to obey the SW model where the coherent rotation dominates, and the mechanism is the magnetization rotation of each grain²³. In this case the coercivity may be used to monitor relative changes in the MAE, and therefore one can derive the MAE from the angular dependence of the magnetization curves $M(H)$ using^{24, 25}

$$MAE = \frac{\int_0^{M_s} (HdM_{\gamma_1} - HdM_{\gamma_2})}{\sin^2(\gamma_1 - \gamma_2)} \quad (1)$$

where H is the applied magnetic field, $\gamma_1 = 0^\circ$ and $\gamma_2 = 60^\circ$, and M_s is the total magnetic moment estimated at the saturation field using

$$M_s = m_{L,Co} + m_{\text{seff},Co} \quad (2)$$

where $m_{L,Co}$ and $m_{\text{seff},Co}$ are the orbital and spin magnetic moments of Co, respectively. Using the sum rules analysis, we evaluated the Co 3d total effective magnetic moment as $M_s \sim 1.4 \mu_B / \text{atom}$, which does not vary much with the OVF.

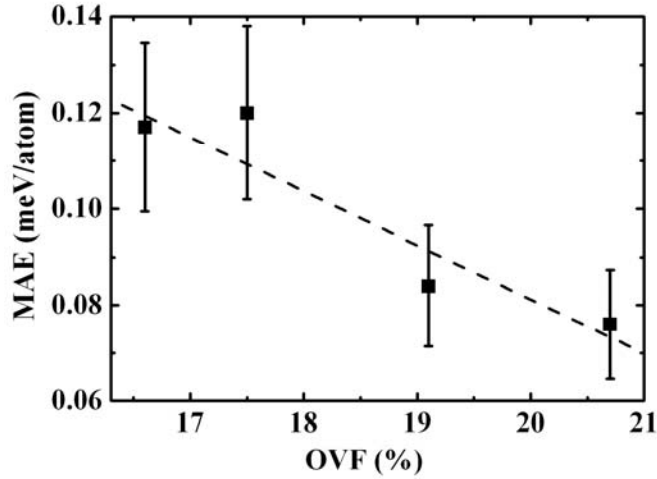


FIG. 6: Co MAE (black squares) vs. OVF determined from Eqs. (1) and (2). Dashed line is a linear fit to the MAE data.

A strong dependence of the MAE on OVF is observed in Fig. 6 with a trend similar to that of the H_C measured by VSM as well as the Co L edges XMCD. The

value of the Co MAE decreases from 0.117 meV/atom to 0.076 meV/atom as the OVF increases from 16.6% to 20.7%, which is consistent with the decreasing trend of K_{1g} with increasing OVF observed in Ref. 5.

V. Conclusions

We have measured for the first time the element-specific hysteresis loops for $\text{Co}_{80}\text{Pt}_{20}$ thin films with perpendicular magnetization using angle-dependent XMCD at the $\text{Co}L_{2,3}$ edges. The magnetization-reversal mechanism of these samples demonstrated to be dominated by the magnetization rotation of each isolated grain, and the Co MAE is evaluated from the angular dependence of $M(H)$ in the XMCD hysteresis loops accordingly. When the OVF increases from 16.6% to 20.7%, the Co MAE decreases from 0.117 meV/atom to 0.076 meV/atom. This work has shown that $\text{Co}_{80}\text{Pt}_{20}$ thin films with 19.1%-20.7% OVF possess the most compromised conditions for high-density perpendicular magnetic recording: namely, sufficiently small and exchange-decoupled magnetic grains with a large MAE and negative nucleation field.

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