Characterization of Oxide Materials for Exchange Decoupling in Perpendicular Thin Film Media

Vincent M. Sokalski¹, Jian-Gang Zhu², Fellow, IEEE, and David E. Laughlin¹

¹Department of Materials Science & Engineering, DSSC, Carnegie Mellon University, Pittsburgh, PA 15213 USA ²Department of Electrical & Computer Engineering, DSSC, Carnegie Mellon University, Pittsburgh, PA 15213 USA

We report on continued measurements using an experimental model system to quantify intergranular exchange coupling in CoCrPtoxide perpendicular magnetic recording media. A thin film multi-layered structure comprising a high coercivity CoPt unicrystal layer separated from a low coercivity CoPt layer by a thin oxide interlayer is used to model the vertically aligned grains separated by oxide boundaries in CoCrPt-oxide media. Exchange coupling is measured by field shifts of the minor loop from the low coercivity layer. Results on coupling energy as a function of interlayer thickness are presented for several different oxides. Additional measurements are presented to understand the possible role of the added alloying elements, Cr and Mn, to the magnetic grains in terms of intergranular exchange coupling.

Index Terms—Intergranular exchange coupling, perpendicular magnetic recording.

I. INTRODUCTION

RESENT perpendicular thin film media contains a recording layer comprised of at least two magnetic layers: a CoCrPt-oxide layer with magnetic grains separated by oxide grain boundaries and a continuous CoCrPt capping layer with no oxide material. In order to increase the areal recording density of perpendicular media, the grain size must be made smaller in the CoCrPt-oxide layer to maintain a sufficient signal to noise ratio (SNR). The oxide material in this layer should fully decouple the neighboring magnetic grains while the capping layer provides spatially uniform intergranular exchange coupling [1]. It is suggested that significant intergranular exchange coupling actually does occur in the CoCrPt-oxide layer, particularly for media with very small grain sizes [2], [3]. For this reason, it's critical to understand how the exchange coupling between neighboring grains varies with the thickness and type of oxide in the grain boundary separating them.

II. EXPERIMENTAL PROCEDURE

In previous work, it was shown that an experimental model system could be used to assess the ability of various oxides to exchange decouple neighboring layers of CoPt. [4]. In this paper, we report on continued work using this experimental method and extend the model to include various additional oxides as well as elemental interlayers. The model system consists of two ferromagnetic $Co_{84}Pt_{16}$ layers separated by an oxide interlayer grown epitaxially on an MgO(110) single crystal substrate. Films were prepared by RF sputtering using a Leybold Heraeus Z400 sputtering system maintained at a base pressure of 2×10^{-7} Torr and an argon sputtering pressure of 5 mTorr. A schematic of the epitaxial relationships and thin film structure is shown in Fig. 1. The orientation relationships can be summarized as follows:

$CoPt(10\overline{1}0) ||Cr(112)||MgO(110)$ $CoPt[0001]||Cr[1\overline{1}0]||MgO[001].$

These epitaxial relationships have been used extensively in the literature and are described in detail in other work [5], [6]. A 60 nm thick Cr underlayer is deposited with the substrate heated to 400 °C. The increased temperature provides the energy necessary for the Cr to grow epitaxially with (112) orientation. After cooling to room temperature, the remaining CoPt (24 nm)/oxide (δ_{ox})/CoPt (8 nm) layers are deposited. The 24 nm bottom layer of CoPt is hexagonal close-packed (HCP) and grows with (1010) texture. This layer is known as a "unicrystal," which has a single c axis orientation variant lying in the plane of the film. The top layer, which is grown on the usually amorphous oxide interlayer, has no particularly strong lattice orientation and, consequently, a relatively low coercivity.

These epitaxial relationships were verified by X-ray diffraction (XRD) in a previous report [4] and the cross section electron diffraction pattern (Fig. 2(A)) viewed along the MgO[001] direction. The predicted pattern indicates the corresponding layer for each diffraction spot. Most important is the 6-fold CoPt pattern associated with a [0001] zone axis, which indicates that the c axis is lying in the film plane. Also noteworthy is the pattern associated with the Cr layer. The Cr(112) layer grows with two possible orientation variants (Fig. 1(A)) giving rise to two separate Cr diffraction patterns when viewed in cross section along the $Cr[1\overline{10}]$ zone axis. This also leads to a series of double diffraction spots indicated by an"X" in the simulated pattern. The superimposed diffraction patterns from the twin variants combined with their double diffraction spots form a pattern that looks exactly like a Body-Centered Cubic (BCC) [110] pattern, but with a significantly larger lattice parameter. This unique pattern is nonetheless a result of the twin variants which do have the expected 2.88 Å lattice parameter of BCC Cr. The magnetic anisotropy of the CoPt unicrystal is also verified by the in-plane hysteresis loops measured along the orthogonal MgO [001] (easy axis) and MgO[110] (hard axis) directions as shown in Fig. 3(A). A thicker 50 nm unicrystal layer without a soft

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Fig. 1. (A) Schematic CoPt/Cr/MgO epitaxial relationships showing the two possible orientation variants of Cr. (B) Thin film structure used to measure coupling through an oxide boundary.



Fig. 2. (A) Cross section electron DP(left) and predicted pattern (right). The rectangular outlines distinguish patterns from the two possible Cr variants with their resulting double diffractions spots marked 'X'. (B) TEM cross section image for a 1.5 nm SiO_x interlayer [4].



Fig. 3. (A) In-plane hysteresis loops of CoPt Unicrystal measured along the easy & hard axes [4]. (B) Measured major and minor hysteresis loops taken from samples with varying thickness of CrO_X interlayers.

over layer was used for these particular hysteresis loops. The coercivity of the easy axis loop is around 1 kOe even though the anisotropy field, H_K , is much greater as seen by the hard axis loop.

A representative cross section transmission electron microscope (TEM) micrograph (Fig. 2(B)) shows both CoPt layers and a SiO_X layer separating them. The SiO_x interlayer thickness of 1.5 nm is verified and shown to be reasonably smooth with no visible discontinuities.

III. RESULTS AND DISCUSSION

Fig. 3(B) shows a set of M-H loops from this multilayer structure for a CrO_x interlayer of varying thickness with the applied field along the easy axis. The exchange coupling field is determined by measuring the field shift, H_{shift} , of the minor hysteresis loops taken from the low coercivity top CoPt. The interlayer coupling energy density is calculated according to

$$\sigma = H_{\rm shift} M_{S,\rm soft} \delta_{\rm soft} \tag{1}$$

where $M_{s,soft}$ (1100 emu/cm³) and δ_{soft} (8 nm) are the saturation magnetization and thickness of the top CoPt layer. Additionally, for each oxide there is a thickness at which point it is no longer possible to observe separate switching of the two layers. However, the coupling energy density can still be calculated from a decrease in the reversal field of the major loop following the method of van der Heijden *et al.* [7] and is described in a previous publication [4].

The interlayer coupling energy density as a function of oxide thickness is shown in Fig. 4 for several different oxides: SiO_2 , TiO_2 , MgO, Cr_2O_3 , and Y_2O_3 . The compositions listed are those of the sputtering target. The actual oxygen content of the film itself may be quite different. In all cases the coupling is ferromagnetic in nature and the trends can be fit well to the following thickness dependence

$$\sigma = A \exp\left(-\frac{\delta_{\text{ox}}}{B}\right) + C.$$
 (2)

While magnetostatic "orange peel" coupling between the two CoPt layers can explain the non-zero offset at large thickness [8], [9], the sharp exponential increase at lower thickness cannot be explained by this mechanism for any reasonable interfacial roughness parameters. It is also known that ferromagnetic exchange coupling can be mediated by stray fields associated with the formation of domains in both layers [10]. However, the bottom unicrystal layer is a single domain during the entire switching process of the top CoPt layer. Therefore, domain wall coupling is not a viable mechanism to explain this ferromagnetic coupling either. For these films, the most probably coupling mechanisms are ferromagnetic "pinhole" coupling [7], [11] or indirect exchange coupling [12]. Detection of pinholes experimentally is very difficult and a more rigorous study of the interlayer morphology would be necessary to understand, in detail, their contribution to the interlayer exchange. While the exact mechanism of the exchange remains unclear, it is important to recognize that regardless of the dominant mechanism, the trend is highly dependent on which oxide was used. This observation is likely related to fundamental properties of the CoPt/oxide interface, which vary for different oxides.

The critical thickness, t_c , has been defined here to be the point where the coupling energy density reaches 0.2 ergs/cm². The critical thicknesses for CrO_x , TiO_x , and MgO_x are very similar at a thickness of approximately 12 Å. Yttrium oxide (YO_x) results in the strongest decoupling of the CoPt layers with a critical thickness of 9.5 Å. It is particularly interesting to note the



Fig. 4. Coupling energy density versus oxide interlayer thickness for pure oxide interlayers.



Fig. 5. Coupling energy versus oxide interlayer thickness with and without Cr "dusting" for TiO_X (left) and SiO_X . (right). The markers are measured values whereas the lines are trends fit by (2).

significant exchange coupling ($t_c = 18 \text{ Å}$) that occurs in SiO_x, one of the most predominantly used oxides in real media, even though the layer is seemingly continuous.

However, the grains in present perpendicular media are actually comprised of CoCrPt rather than the CoPt used in these experiments. There have been a number of experimental studies which show that Cr segregates to the grain boundaries in real media [13]-[15] and it has long been suggested that a Cr-rich CoCr grain boundary could suppress intergranular exchange coupling [16]. To incorporate this effect into our experimental model system, we have performed an additional set of experiments where a very thin (1-1.5 Å) layer of Cr was deposited before and after deposition of the oxide layer. This interfacial "dusting" was performed for both SiO_x and TiO_x interlayers, the two most frequently used intergranular materials for present perpendicular recording media. The effect of Cr "dusting" is shown in Fig. 5. For the case of SiO_x, the Cr effect is substantial causing the trend to shift by almost 8 Å. This is much greater than the added thickness of chromium. While the shift was much less significant for the case of TiO_x , it is interesting to note that SiO_x and TiO_x have a very different critical thickness before Cr dusting, but a nearly equal one after.

In longitudinal media, the segregation of Cr to the grain boundaries was the dominant mechanism by which the magnetic grains were decoupled and the use of an oxide was not necessary [17]. In addition to Cr, other alloying elements, such as Mn, had been tried experimentally for the same purpose [18]. However, Cr had always been sufficient to decouple the magnetic grains and other alloying elements were not needed. As the grain boundaries continue to be made smaller in finer grained media, the role of a segregating material may



Fig. 6. Comparison of coupling energy density versus interlayer thickness for pure Cr and Mn interlayers using cobalt as the low coercivity top layer. Inset hysteresis loops are for 10 Å and 30 Å Mn layers.

become more important in perpendicular media to enhance decoupling. Motivated by this idea of added "segregating" elements, preliminary work has been done using Cr and Mn as pure elemental interlayers to compare their decoupling ability independent of the oxide material. However, both Cr and Mn can grow epitaxially on the CoPt unicrystal and induce oriented unicrystal growth of the top CoPt layer. In such a case, both the top and bottom layers would have a similar coercivity and observation of separate switching would be difficult. This problem is easily solved by replacing the top layer of high anisotropy CoPt with significantly lower coercivity pure Co with $M_S = 1414$ emu/cm³. The results are shown in Fig. 6. The critical thickness of pure Cr compared to pure Mn is substantial. While Mn has a critical thickness comparable to that of several of the oxides at 12 Å, Cr has a critical thickness of 7.5 Å. This is an improvement over even the best oxide, YO_x .

It is possible that the enhanced decoupling associated with using Cr (either as a pure element or as a "dusting" layer) is related to diffusion of Cr into the CoPt layers. Interfacial diffusion between the two layers could result in the formation of a thin CoCrPt layer with enough Cr (more than 25 at%) to make the layer paramagnetic. It is reasonable to expect that such added paramagnetic material would further decouple the magnetic layers.

Certainly, the growth mechanism in the planar deposition model presented here is quite different from the columnar growth of grains with oxide boundaries in real media. Nonetheless, if the exchange coupling is related to properties of the CoPt/oxide interface, then the results are still applicable to the behavior of real media. A high CoPt/oxide interfacial energy may result in the formation of a less uniform interlayer as a consequence of the non-wetting behavior of high energy interfaces. Such interfacial energy may play a critical role in the formation of uniform and continuous boundaries in real media. If the dependence shown in Figs. 4-6 can be applied to magnetic grains in real media made, then any adjacent grains separated by a boundary less than the critical thickness would experience an exponential increase in intergranular exchange coupling. Furthermore, a distribution in grain boundary thickness below this critical thickness would result in a large variation in exchange coupling energy throughout the media. Micromagnetic modeling calculations based on the experimental work presented here have indicated that there is a profound influence of "turning on" intergranular exchange coupling on the signal to noise ratio [19]. In particular, it has been shown that if non-uniformly distributed exchange coupling is considered, there is no gain in SNR when the grain size is decreased from 9 nm to 6 nm as long as experimental grain boundary thickness distributions are considered.

IV. CONCLUSION

We have measured the exchange coupling strength of several different oxide materials and studied the possible role of Cr and Mn as added alloying elements to the magnetic grains. In all cases, the interlayer material shows an exponential dependence on thickness below a critical value. This critical thickness is a strong function of the oxide material and can be decreased by interfacial "dusting" of chromium, particularly for SiO_x. While there are certainly differences between the formation mechanisms in the model system presented here and the formation process of grain boundaries in real media, the results nonetheless provide a quantitative way to evaluate the problem of intergranular exchange coupling. Controlling intergranular exchange coupling in real media is a necessary challenge that needs to be met in order to improve the recording properties of current perpendicular media. One solution is to process the media in such a way to minimize the distribution in grain boundary thickness and, as such, minimize the distribution in exchange coupling energy throughout the film. A more promising approach, as suggested by this paper, is to engineer the grain boundary material to have a low critical thickness. Such improvements may be realized by a combination of selecting different oxide materials and/or implementing added alloying elements that may segregate to the grain boundaries and further decouple the magnetic grains. In either case, the experimental results presented here provide a starting point for systematically developing improved boundaries in perpendicular magnetic recording media.

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