Study of Magnetic Easy Axis 3-D Arrangement in $L1_0$ CoPt(111)/Pt(111)/MgO(100) Tilted System for Perpendicular Recording

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We report a study of angular magnetic properties of high-anisotropy $L1_0$ CoPt (111) films having a tilted magnetic easy axis configuration without an oblique-grain microstructure. In particular, we investigated the field dependence of remanent magnetization while rotating the magnetic field inside three intersecting planes. The out-of-plane tilting of the $L1_0$ c-axis (the easy axis of the tetragonal cell) was induced by using a Pt (111) underlayer deposited onto a single-crystal MgO substrate in a conventional frontal pulsed laser deposition (PLD). The observed behavior is consistent with the presence of four easy axes with mutually orthogonal in-plane projections, symmetrically tilted at 36° with respect to the film plane. Such a system can be used, like a common single-axis tilted medium, to record information in perpendicular mode, lowering the writing field to approximately 75% of the value along the easy direction, while still maintaining the high thermal stability typical of the $L1_0$ alloy. Moreover, the in-plane charge compensation arising from this easy axis arrangement when a perpendicular writing field is applied may favor a media noise reduction and better performance with respect to a single-axis tilted system.

***Index Terms*—EDXD, $L1_0$ CoPt (111), PLD, tilted magnetic recording.**

I. INTRODUCTION

SIGNIFICANT progress in hard disk technology, in terms of increased areal density of the recording media, is currently limited by the tradeoff among writability [1], thermal stability [2], and medium signal-to-noise ratio [3], the so-called recording *trilemma* [4].

The perpendicular magnetic recording mode [5] has been proposed to break such a trilemma. However, there are still some open issues for it to become competitive. Among them, very critical is the high sensitivity of the media noise on the switching field distribution (SFD) [6] which, according to the Stoner–Wohlfarth model [7], is quite broad even when the easy axis distribution (EAD) is very narrow (e.g., 1 degree). Moreover, the EAD depends on sample microstructure and it has always a finite width unless using an epitaxial film.

In conventional industrial deposition equipment, the rigorous control of film epitaxy is expensive and difficult to obtain. Therefore, to overcome such a limitation, the idea of using tilted media was recently reintroduced and such systems have been suggested as leading candidates for extending hard disk drive areal density beyond 1 Tb/in^2 [8]. They require the magnetization easy axis of the grains to be uniformly tilted at an optimal angle of 45° with respect to the surface normal: in such a way, the sensitivity of SFD to EAD is highly reduced and less demanding deposition conditions are required. Furthermore, the 45° tilted direction is associated with the minimum switching field value, allowing higher magnetic anisotropy materials to be used, such as the tetragonal $L1_0$ phase of the MPt (M = Co, Fe) alloys [9].

Several theoretical works have been recently presented on the advantages of using tilted media for perpendicular recording and different media designs have been analyzed [6], [8], [10]–[12]. However, only a few attempts have been carried out up to now to fabricate such media and most of them employed a growth process in oblique direction [13], [14], even though it is well known that the oblique grains microstructure can negatively influence the magnetic properties due to the increased roughness and stress and shape anisotropy effects [15].

For the high-anisotropy face-centered tetragonal fct $L1_0$ alloy, a better approach is to control the growth direction by a suitable underlayer such as Pt (111). Indeed, crystallographic considerations indicate that the [001] axis (the easy axis of the tetragonal cell) of (111) oriented $L1_0$ CoPt films is about 36° tilted from substrate surface, i.e., very close to the ideal 45° angle suggested in the literature, and such films can then be considered natural candidates for tilted media. Studies on the magnetic properties of such a system, deposited onto different underlayers and substrates, have already been presented in the literature [16]–[18], however, the global angular dependence of magnetization has not been clarified, since measurements were mostly performed in longitudinal and perpendicular directions. By such measurements, a complete understanding of the easy axis geometry is not achievable and the geometrical picture of easy axis 3-D arrangement can only be hypothesized.

II. EXPERIMENT

$Co_x Pt_{1-x}$ thin films (x $\sim$ 0.5, thickness $\sigma \sim 15$ nm) were deposited in HV conditions ($P_{\rm dep} = 10^{-7}$ mbar) by PLD onto a polished single-crystal MgO (100) substrate at a laser fluence $F = 3$ J/cm^2 and $T_{\rm dep} = 873$ K from a CoPt rotating target formed by Co and Pt sectors. The laser beam formed an angle of 45° with the target surface and target and substrate were placed in a frontal geometry at a distance of 55 mm. A very thin and highly oriented Pt (111) underlayer ($\sigma \sim 4$ nm) was used to favor the epitaxial growth of the magnetic layer along the [111] direction. On the basis of our previous work [19], the Pt (111) orientation was obtained by depositing at $F = 3$ J/cm^2 and $T_{\rm dep} = 573$ K, where only this growth direction was obtained.

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After the deposition, the samples have been annealed *in situ* in order to favor the transition from the magnetically soft fcc to the magnetically hard fct $L1_0$ CoPt phase. Here, we focus on results obtained by annealing at $T_{\rm ann} = 873$ K for 3 h. Structural properties and their influence on the magnetic behavior were studied by combining X-ray diffraction (in the energy dispersive mode) and magnetization measurements.

The energy dispersive (ED) X-ray diffraction measurements were performed by a noncommercial equipment [20], [21] based on the use of a polychromatic ("white") primary X-ray beam, produced by a W anode, and of an ultra-pure Ge solid-state device detector, which allows performing the energy scan of the diffracted photons. In such a way, the reciprocal space scan (q-scan, where q is the normalized momentum transfer magnitude), necessary to collect the diffraction pattern, is carried out electronically, rather than mechanically as in the conventional (i.e., angular dispersive) X-ray diffraction mode. The main advantage of the ED mode is that the experimental setup can be kept fixed during the acquisition of the diffraction patterns, simplifying the experimental geometry and preventing systematic angular errors, as well as possible misalignments. In particular, as far as structural analysis is concerned, this technique provides a fast recording of Bragg peaks and their rocking curves (RCs) since, in the ED mode, the whole pattern is obtained simultaneously at each q-value. The RC analysis provides the statistical distribution of the film domain's orientation, i.e., the degree of epitaxy along the growth direction. To collect them, a series of diffraction patterns is acquired as a function of an asymmetry parameter (rocking angle) $\beta = (\vartheta_i - \vartheta_f)/2$, where ϑ_i and ϑ_f are the initial (incidence) and final (deflection) angles with respect to the film surface and $\vartheta_i + \vartheta_f = 2\vartheta$ is kept constant [22].

The magnetic measurements were taken by a commercial vectorial VSM magnetometer (model 10—ADE Technologies), equipped with a rotating electromagnet for studying the angular dependence of magnetization.

III. Results and Discussion

The ED X-ray diffraction experiments revealed the presence of the CoPt (111) reflection at $q = 2.875(5)$ Å^{-1}, confirming a preferential growth along the (111) direction, no other Bragg reflections being detected in the q-range investigated. Moreover, the X-ray data showed that the Pt underlayer is grown along the (111) orientation, as revealed by the presence at $q = 2.775(5)$ Å^{-1} of the Pt (111) cubic phase reflection. The residual peak at $q = 2.982$ Å^{-1} was attributed to the MgO monocrystalline substrate (200) reflection. This information was deduced by the analysis of the sequence of diffraction patterns collected as a function of the scattering parameter q and at different asymmetry values β and shown in Fig. 1. Apart from the Bragg reflections, in the empty circle two peaks not corresponding to diffraction signal are evidenced. These features, the so-called detector echoes, are due to a statistical effect produced by the detector in correspondence to the most intense peak (the MgO (200) substrate reflection) and are related to the photoemission of a core electron by the Ge single crystal accomplishing the photon detection. However, preliminary tests were performed in order to individuate an appropriate experimental scattering angle locating the echoes in a q-region (2.2 Å$^{-1} < q <$ 2.3 Å^{-1}) where no diffraction signal from the sample was present.

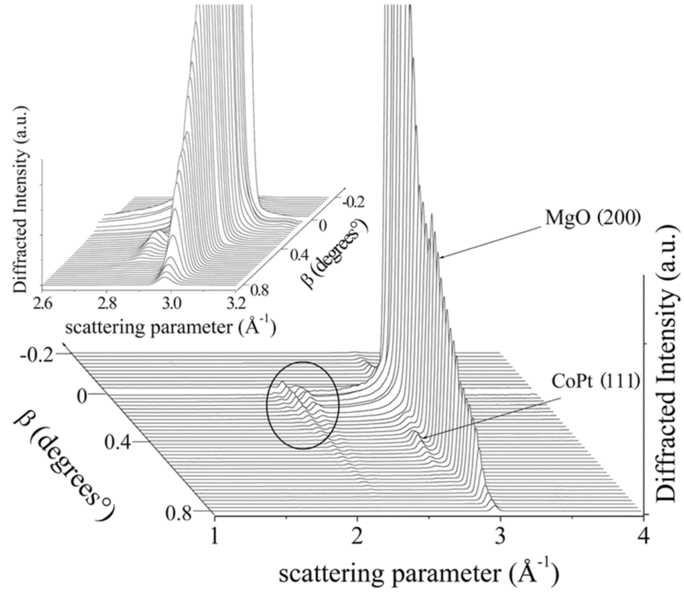

Fig. 1. Sequence of EDXD patterns collected as a function of the scattering parameter q and of the asymmetry parameter β. In the inset, a highlight of the region of interest is shown to evidence the rocking curve of the CoPt (111) reflection.

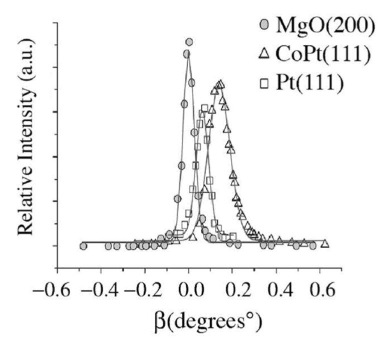

Fig. 2. Rocking curve analysis of the CoPt (111) (—Δ—) and Pt (111) (—□—) reflections. As it can be seen, the FWHM obtained by a Gaussian fit of the experimental data are comparable to that of the MgO (200) monocrystalline substrate (— • —) indicating a highly epitaxial growth of the films. A slight mismatch is also visible by the shift of the Gaussian peak maxima with respect to the MgO (200) substrate.

In the inset of Fig. 1, a highlight of the q-region of interest (2.6 Å$^{-1} < q <$ 3.2 Å^{-1}) is shown in order to evidence the presence of the above-mentioned diffraction peaks. The rocking curve of the CoPt (111) reflection was deduced by an accurate analysis of the relative intensities of the diffraction pattern peaks (Fig. 2). The RCs full-width at half-maximum (FWHM) resulted to be 0.07° and 0.09°, for the Pt (111) and the CoPt (111) reflections, respectively. These values are very close to the FWHM of the MgO (200) reflection rocking curve (FWHM = 0.05°). This is an indication of the high epitaxy degree obtained for both the underlayer and the CoPt film along the growth direction. Moreover, a mismatch of 0.06° and 0.14° was deduced for MgO/Pt and MgO/CoPt layers, respectively.

Since the CoPt film is highly oriented along the [111] direction and the (111) reflection is common to cubic and tetragonal phases [23], it is not possible by X-ray diffraction analysis only, to verify to which extent the phase transformation fcc-fct has

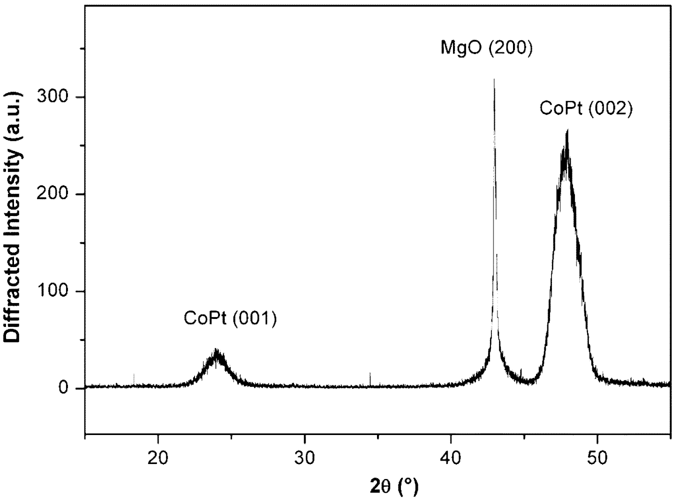

Fig. 3. X-ray (Cu K_α) $\theta - 2\theta$ diffraction pattern (K_a of a CoPt thin film grown at $T_{dep} = 873$ K onto an MgO (100) substrate and post-annealed at $T_{ann} = 873$ K for 3 h. The superlattice (001) reflection of the fct CoPt phase is clearly visible ($2\theta \sim 24.1^\circ$) indicating the appearance of the tetragonal phase.

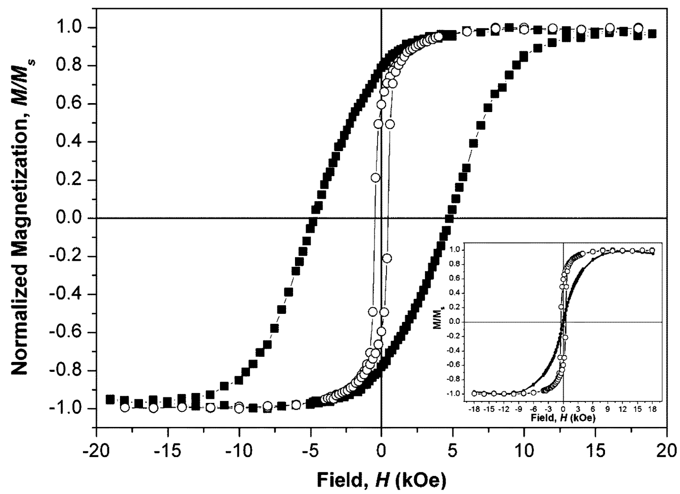

Fig. 4. Comparison between normalized (M/M_s) hysteresis loops of the as-deposited ($- \circ -, H_c \sim 0.46$ kOe) and the annealed samples ($-\blacksquare-, H_c \sim 4.8$ kOe) measured along the easy axis. Inset: Comparison between normalized in-plane ($- \circ -$) and out-of-plane ($- \bullet -$) hysteresis loops of the as-deposited sample.

proceeded (a spectrum collected tilting the sample plane to 36° would be necessary to observe reflections from the 001 planes, which was not possible due to the very low film thickness in comparison to the substrate).

The presence of a magnetically hard phase after the heat treatment was however demonstrated since 1) the same deposition process and annealing procedure were applied to grow CoPt films with (001) orientation and evidence of fct phase formation (appearance of 001 superlattice reflection, Fig. 3) were obtained and 2) the strong increase of coercivity H_c from 0.46 kOe (as-deposited sample) to about 4.8 kOe in the 3h-annealed sample is compatible with fct phase formation (Fig. 4). Moreover, the high deposition temperature (873 K) and the prolonged high-temperature annealing (873 K, 3 h) guaranteed that in our samples any elemental local Co or Pt segregation, if produced by the deposition process, would be totally annihilated. Pt and Co local segregation was invoked, for example, as the origin of a perpendicular anisotropy appearance in $CoPt_3$ metastable alloys deposited at lower temperature ($\sim$400 K) with a consequent increase of the coercive field up to a maximum of 2400 Oe [24]–[27].

Room temperature *angular remanence measurements* (ARM) were carried out in order to determine the geometrical arrangement of the easy axes in the film. They were performed by measuring the remanent magnetization M_r while rotating a field $H = 19$ kOe, from 0° to 360°, in the (110), ($\bar{1}10$), and (001) MgO planes (the two out-of-plane ARM $_{(110)}$ and $\mathrm{ARM}_{(\bar{1}10)}$, and the in-plane $\mathrm{ARM}_{(001)}$, respectively). The spatial position of the highest squareness (M_r/M_s) values coincides exactly with the easy axis directions.

The analysis of the ARM curves (for clarity, only the 0°–180° section is reported in Fig. 5(a) and(b), the rest being totally symmetric), gave evidence of four out-of-plane maxima at $\alpha \sim 36^\circ + \pi$ and $144^\circ + \pi$ within both the (110) and the ($\bar{1}10$) planes, and four in-plane maxima at $\gamma \sim 45^\circ + \pi/2$, respectively. Moreover, coercivity maxima ($\sim$4.8 kOe) were observed when the field was applied along each of the direction of ARM maxima (Fig. 6). No secondary ARM maxima have been observed, ruling out the possibility of domain structures causing some local remanence maxima also along the hard axes. Hence the angular position of the largest squareness (M_r/M_s) values identifies the easy axis directions, that is, four out-of-plane 36° tilted easy axes, whose in-plane projections are along the [110] direction of MgO [Fig. 5(c)].

This particular easy axis arrangement is consistent with the presence of the fct phase, being observed only in annealed samples. Actually, the as-deposited sample that contains only the cubic isotropic phase showed an in-plane easy axis due to the dominant shape anisotropy contribution (inset of Fig. 4). In our samples, the two in-plane orientation variants that form when a Pt (111) layer grows onto an MgO (100) surface [Fig. 7(a)] [28] can cause the formation of four different in-plane orientation variants of the CoPt (111) films as schematized in the picture [Fig. 7(b)].

The remanence coercivity H_r deduced from the DCD curves (plotted in the inset of Fig. 6), which allows the irreversible (or switching) component to be measured, indicated, as expected, an H_r reduction in perpendicular ($\perp$) direction, being $H_{r,\perp}(\sim 4.8$ kOe) about the 75% of $H_{r,\mathrm{EA}}(\sim 6.4$ kOe) measured along the easy axes. This means that the system can behave like a common single-axis tilted medium to record information in perpendicular mode at a sensibly lower writing field. Moreover, from a technical point of view, we believe that, in agreement with the theoretical work of Guan and Zhu [12] on the advanced properties of a bicrystal structure where two easy axes are oriented at ±45° with respect to the perpendicular direction in the cross-track plane, the observed 4 tilted easy axis arrangement could eliminate bit charge edges by compensation when the writing field is normal to the surface. As a matter of fact, using our preparation procedure, magnetic bits could be obtained with four easy axes arranged in space as following: two ±36° easy axes in the cross track plane, similar to the ideal system suggested by Guan and Zhu, and other two ±36° easy axes in the

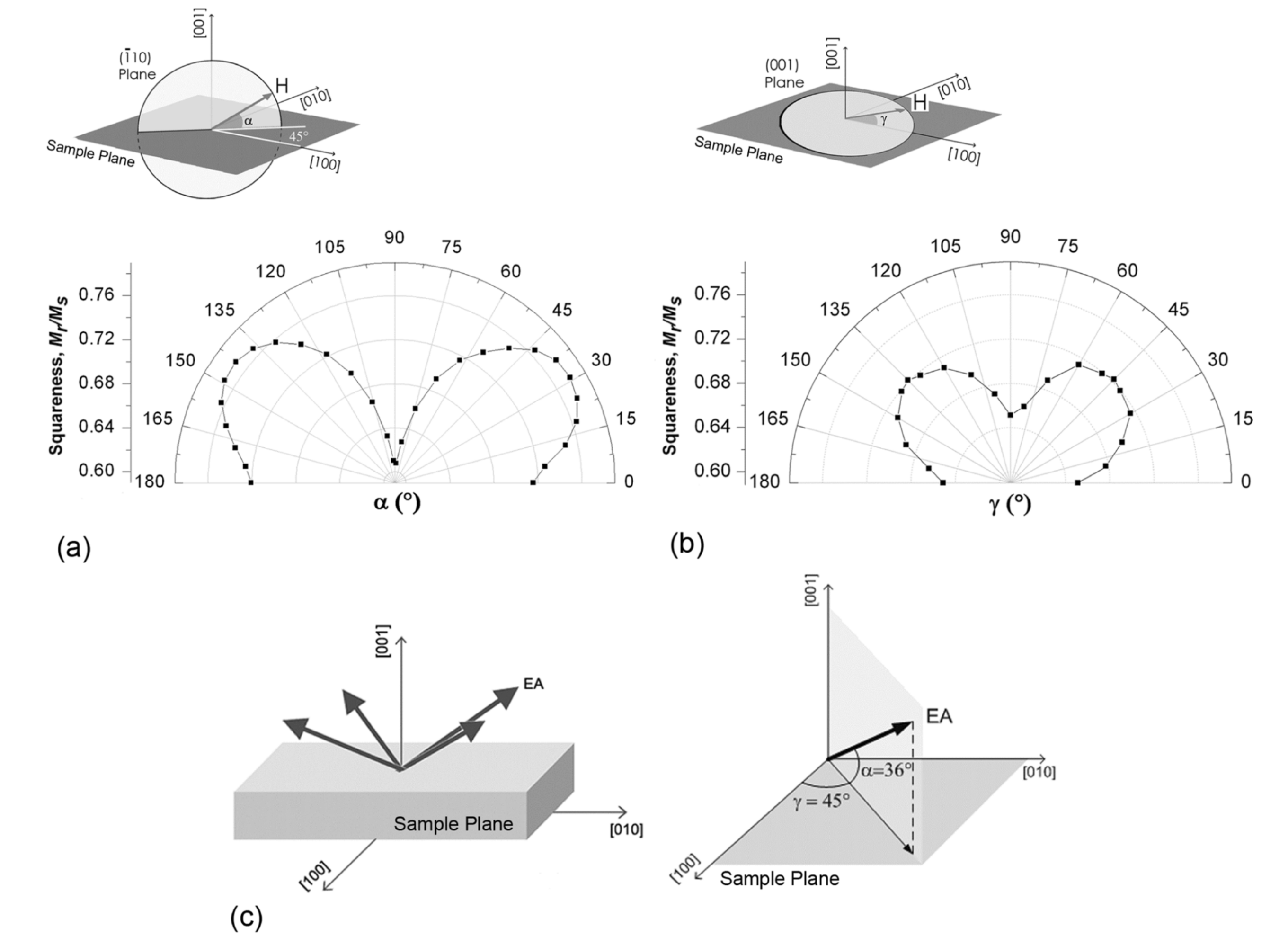

Fig. 5. ARM curves in the (a) $(\bar{1}10)$ and (b) (001) planes of the MgO substrate, whose crystallographic axes constitute the spatial reference system. The ARM in the (110) plane is not reported, being identical to the ARM in the $(\bar{1}10)$ plane. (c) Schematic illustration of the four tilted easy axis (EA) model.

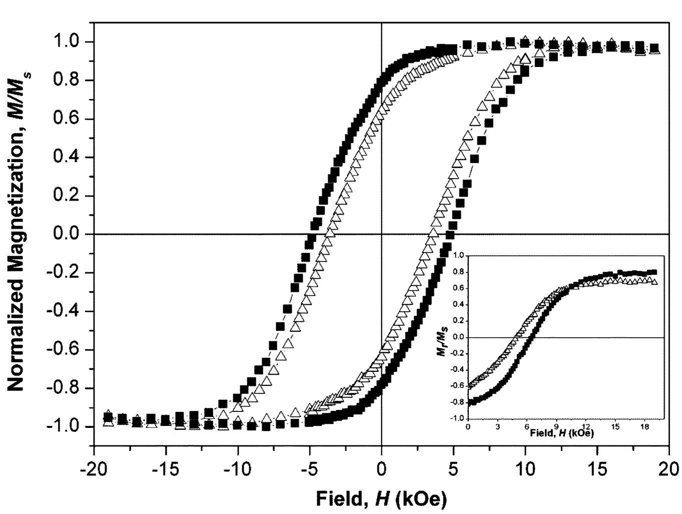

Fig. 6. Normalized (M/M_s) hysteresis loops measured along the easy axes (−■−, $H_c \sim 4.8$ kOe) and the surface normal (−Δ−, $H_c \sim 3.6$ kOe) directions. Inset: Normalized remanence (M_r/M_s) DCD curves measured along the easy axis (−■−) and the surface normal (−Δ−) directions.

down track plane (perpendicular to the cross-track one), which can give rise to an identical effect. This bit edge charge compensation favors a media noise reduction and better performances may be achieved with respect to the most common single-axis tilted system.

IV. Conclusion

We presented the investigation of structural and magnetic properties of a magnetically tilted system without a tilted grains microstructure. In our system, the 3-D angular dependence of magnetization, studied measuring magnetization inside three intersecting planes, was explained in terms of a 4 tilted easy axis arrangement. Such arrangement was never reported in the literature before, even in similarly behaving systems, since only perpendicular and longitudinal magnetization was usually studied. The proposed system could be used, like a common single-axis tilted medium, to record information with a write head in perpendicular geometry, sensibly lowering the necessary writing field $(H_{r,\perp}/H_{r,\mathrm{EA}} \sim 0.75)$ even if employing high-anisotropy alloys such as the $L1_0$ CoPt, needed to guarantee a high medium thermal stability. Moreover, we believe that, in agreement with theoretical predictions, the in-plane charge compensation may favor a media noise reduction and better performances with respect to a single-axis tilted system.

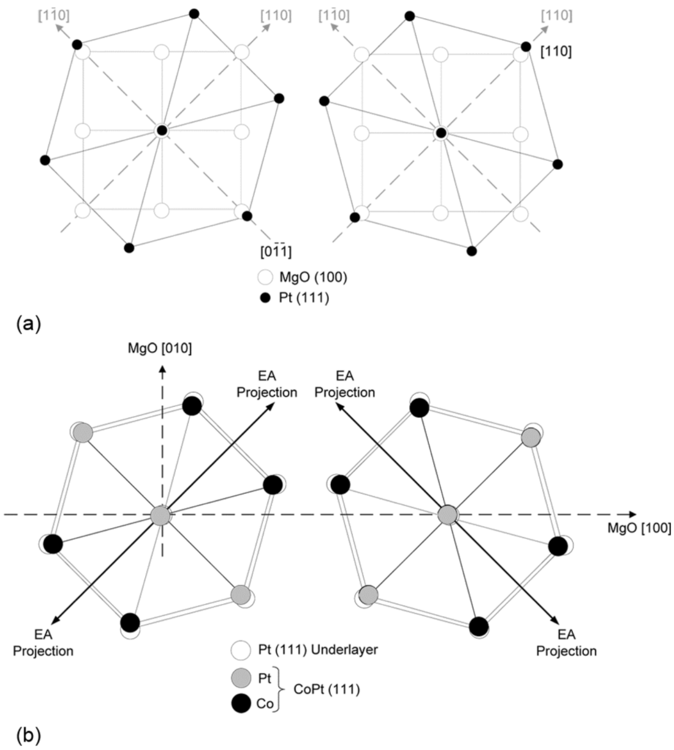

Fig. 7. Schematic representations of the atomic matching between (a) Pt (111) and MgO (100) as reported in [28], and (b) fct CoPt (111) and Pt (111) as we suggested. The two shown Pt in-plane orientation variants ([110])(111)Pt// [110](100) MgO and [110](111)//[1$\bar{1}$0] (100) MgO) may be the origin of the observed 4 tilted easy axis arrangement.

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REFERENCES

[1] R. L. Comstock, "Review: Modern magnetic materials in data storage," *J. Mater. Sci.: Mater. Electr.*, vol. 13, pp. 509–523, Sep. 2002.
[2] S. H. Charap, P. L. Lu, and Y. He, "Thermal stability of recorded information at high density," *IEEE Trans. Magn.*, vol. 33, no. 1, pp. 978–983, Jan. 1997.
[3] H. N. Bertram and M. Williams, "SNR and density limit estimates: A comparison of longitudinal and perpendicular recording," *IEEE Trans. Magn.*, vol. 36, no. 1, pp. 4–9, Jan. 2000.
[4] H. J. Richter and A. Y. Dobin, "Angle effects at high-density magnetic recording," *J. Magn. Magn. Mater.*, vol. 287, pp. 41–50, Feb. 2005.
[5] S. Iwashaki and Y. Nakamura, "An analysis for the magnetization mode for high density magnetic recording," *IEEE Trans. Magn.*, vol. MAG-13, no. 5, pp. 1272–1277, Sep. 1977.
[6] Y. Y. Zou, J. P. Wang, C. H. Hee, and T. C. Chong, "Tilted media in a perpendicular recording system for high areal density recording," *Appl. Phys. Lett.*, vol. 82, no. 15, pp. 2473–2475, Apr. 2003.
[7] E. Stoner and E. Wohlfarth, "A mechanism of magnetic hysteresis in heterogeneous alloys," *IEEE Trans. Magn.*, vol. 27, no. 4, pp. 3475–3518, Jul. 1991.
[8] J. P. Wang, "Tilting for the top," *Nature Mater.*, vol. 4, pp. 191–192, Mar. 2005.
[9] D. Weller, A. Moser, L. Folks, M. E. Best, W. Lee, M. F. Toney, M. Schwickert, J. U. Thiele, and M. F. Doerner, "High K_u materials approach to 100 Gbits/in^2," *IEEE Trans. Magn.*, vol. 36, no. 1, pp. 10–15, Jan. 2000.
[10] K. Z. Gao and H. N. Bertram, "Magnetic recording configuration for densities beyond 1 Tb/in^2 and data rates beyond 1 Gb/s," *IEEE Trans. Magn.*, vol. 38, no. 6, pp. 3675–3683, Nov. 2002.
[11] J. P. Wang, Y. Y. Zou, C. H. Hee, T. C. Chong, and Y. F. Zheng, "Approaches to tilted magnetic recording for extremely high areal density," *IEEE Trans. Magn.*, vol. 39, no. 4, pp. 1930–1935, Jul. 2003.
[12] L. Guan and J. G. Zhu, "Bicrystal structure of tilted perpendicular media for ultra-high-density recording," *J. Appl. Phys.*, vol. 93, pp. 7735–7737, May 2003.
[13] K. Tanahashi, Y. Hosoe, and M. Futamoto, "Magnetic anisotropy and microstructure of obliquely evaporated Co/Cr thin films," *J. Magn. Magn. Mater.*, vol. 153, pp. 265–272, Feb. 1996.
[14] Y. F. Zheng, J. P. Wang, and V. Ng, "Control of the tilted orientation of CoCrPtTi thin film media by collimated sputtering," *J. Appl. Phys.*, vol. 91, pp. 8007–8009, May 2002.
[15] T. Klemmer and K. Pelhos, "Seed layer control for tilted magnetic recording media," *Appl. Phys. Lett.*, vol. 88, p. 162507, Apr. 1–3, 2006.
[16] S. E. Park, P. Y. Jung, and K. B. Kim, "Magnetic properties and microstructural analysis of sputtered-deposited and annealed Co-Pt alloys," *J. Appl. Phys.*, vol. 77, pp. 2641–2647, Mar. 1995.
[17] V. Karanasos, I. Panagiotopoulos, and D. Niarchos, "Texture and strain in CoPt/Ag nanocomposite films," *J. Magn. Magn. Mater.*, vol. 249, pp. 471–474, Sept. 2002.
[18] D. Y. Oh and J. K. Park, "Crystallographic texture and angular dependence of coercivity of ordered CoPt thin film," in *Proc. 49th Annu. Conf. Magnetism and Magnetic Materials*, Jacksonville, FL, 2004, pp. 10N105 1–3.
[19] G. Scavia, E. Agostinelli, S. Laureti, G. Varvaro, B. Paci, A. Generosi, V. Rossi Alberini, S. Kaciulis, and A. Mezzi, "Evolution of the Pt layer deposited on MgO(001) by pulsed laser deposition as a function of the deposition parameters: A scanning tunneling microscopy and energy dispersive X-ray diffractometry/reflectometry study," *J. Phys. Chem. B*, vol. 110, pp. 5529–5536, Mar. 2006.
[20] R. Caminiti and V. Rossi Alberini, "The kinetics of phase transition observed by energy dispersive X-ray diffraction," *Int. Rev. Phys. Chem.*, vol. 18, pp. 263–299, 1999.
[21] V. Rossi Albertini, B. Paci, and A. Generosi, "Review: The energy dispersive X-ray reflectometry as a unique laboratory tool to investigate morphological properties of layered systems and devices," *J. Phys. D: Appl. Phys.*, vol. 39, pp. 461–486, 2006.
[22] B. Paci, A. Generosi, V. Rossi Albertini, E. Agostinelli, G. Varvaro, and D. Fiorani, "Structural and morphological characterization by energy dispersive X-ray diffractometry and reflectometry measurements of Cr/Pt bilayer films," *Chem. Mater.*, vol. 16, pp. 292–298, 2004.
[23] A. Cebollada, R. F. C. Farrow, and M. F. Toney, "Structure and magnetic properties of chemically ordered magnetic binary alloys in thin film form," in *Magnetic Nanostructures*, ACP, 2002, pp. 93–122, ch. 3.
[24] M. Maret, M. C. Cadeville, R. Poinsot, A. Herr, E. Beaurepaire, and C. Monier, "Structural order related to the magnetic anisotropy in epitaxial (111) $CoPt_3$ alloy films," *J. Magn. Magn. Mater.*, vol. 166, pp. 45–52, Feb. 1997.
[25] M. Albrecht, M. Maret, A. Maier, F. Treubel, B. Riedlinger, U. Mazur, G. Schantz, and S. Anders, "Perpendicular magnetic anisotropy in $CoPt_3$ (111) films grown on a low energy surface at room temperature," *J. Appl. Phys.*, vol. 91, pp. 8153–8155, May 2002.
[26] P. W. Rooney, A. L. Shapiro, M. Q. Tran, and F. Hellman, "Evidence of a surface-mediated magnetically induced miscibility gap in Co-Pt alloy thin film," *Phys. Rev. Lett.*, vol. 75, pp. 1843–1846, Aug. 1995.
[27] J. P. Hu and P. Lin, "High-coercivity CoPt alloy films grown by sputtering," *IEEE Trans. Magn.*, vol. 32, no. 5, pp. 4096–4098, Sep. 1996.
[28] C. Gate, P. Baules, and E. Snoeck, "Morphology of Pt islands grown on MgO (001)," *J. Cryst. Growth*, vol. 252, pp. 424–432, May 2003.

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