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Influence of the substrate choice on the L1₀ phase formation of post-annealed Pt/Fe and Pt/Ag/Fe thin films

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⁶I. Frantsevich Institute for Problems of Materials Science, National Academy of Sciences of Ukraine, 1-FePt phase after post-annealing or after deposition in thermal expansion coefficients of the metallic film and the substrates (Al₂O₃(0001): 10 × 10⁻⁶ K⁻¹, SrTiO₃(001): 9.4 × 10⁻⁶ K⁻¹, and MgO(001): 5 × 10⁻⁶ K⁻¹ (Ref. 8)).

II. EXPERIMENTAL

Layered films of Pt(15 nm)/Ag(0; 10 nm)/Fe(15 nm) were deposited at room temperature on MgO(001), SrTiO₃(001), and Al₂O₃(0001) single crystalline substrates by DC magnetron sputtering using individual Pt, Ag, and Fe targets. The Ar pressure in the sputtering chamber was adjusted to 0.48 Pa for all depositions. The nominal thicknesses of the layers were evaluated from the calibrated sputtering time (determined from the calibrated deposition rate of each target) and verified by profilometer measurements. Post-annealing of the films up to 1173 K was carried out in flowing N₂ atmosphere with a constant heating rate of 10 K/s. The structure of the films was analyzed with an x-ray diffractometer (XRD) equipped with 2-dimensional (2D) and scintillation detectors using Cu Kα radiation. Composition-time (depth) profiles of post-annealed samples were determined by SNMS, using a low-pressure radio-frequency Ar plasma both as source for ion bombardment and as post-ionization medium. Furthermore, the magnetic properties were measured by superconductive quantum interference device-vibrating sample magnetometry (SQUID-VSM).

III. RESULTS

A. Pt/Fe and Pt/Ag/Fe films on MgO(001)

Fig. 1 shows 2D XRD images of Pt/Fe and Pt/Ag/Fe films sputter-deposited on MgO(001) substrates after post-annealing...
at various temperatures. The appearance of the (001) superstructure peak for both Pt/Fe and Pt/Ag/Fe films was weakly indicated after annealing at 773 K but clearly observed after annealing at 873 K, indicating the onset of the L10-FePt phase formation. With increasing annealing temperature, this reflection increases in intensity and becomes sharper. However, the intensity of the (111) peak is still much higher than the intensity of (001) reflection, indicating the presence of strong (111)-texture. Furthermore, the intensity of the (111) Ag reflection observed for the Pt/Ag/Fe films also becomes more pronounced with increasing temperature. Please note that it is well known that FePt films grown on MgO(001) substrates at elevated temperatures reveal high L10 ordering with pronounced (001) texture. However, in our case, the ordering mechanism is quite different as bilayer Fe/Pt films were deposited at room temperature and then post-annealed. Thus, FePt ordering is initiated in the Fe/Pt bilayer apart from the substrate.

SNMS composition profiles versus sputtering time of Pt/Fe and Pt/Ag/Fe films after post-annealing at 773 K and 873 K are presented in Fig. 2. Please note that the composition profile was calculated assuming a linear dependence of the measured intensities on the elemental concentration. Post-annealing of the Pt/Fe film at 773 K leads to an almost homogeneous intermixing between the Pt and Fe layers. Further increase of the temperature up to 873 K does not change significantly the concentration profile. Post-annealing of the Pt/Ag/Fe film at 773 K also leads to the formation of a homogeneous FePt layer and to a moderate penetration of Ag into the FePt layer with an Ag rich layer on the top surface. Please note that from the individual Fe and Pt layer thicknesses, a composition of Fe57Pt43 is expected.

B. Pt/Fe and Pt/Ag/Fe films on SrTiO3(001)

Fig. 3 shows 2D XRD images of Pt/Fe and Pt/Ag/Fe films sputter-deposited on SrTiO3(001) substrates after post-annealing at temperatures up to 1173 K. Please note that the (100) reflection of the SrTiO3 substrate and the (001) reflection of L10-FePt are superimposed and cannot be easily distinguished. However, the onset of L10 chemical ordering was registered after post-annealing between 773 K and 873 K in both Pt/Fe and Pt/Ag/Fe films. Intensity of superstructure reflection becomes stronger with higher annealing temperatures. It is apparent that after post-annealing at 1073 K and 1173 K, a non-uniform distribution of the (001) peak intensity along the diffraction ring is observed, revealing that some part of the grains are preferentially oriented along the [001] direction. But the strong (111) peak is still present in the XRD images. The SNMS depth profiles of post-annealed films grown on SrTiO3(001) substrates (Fig. 4) are very similar to the
profiles obtained on MgO(001) substrates. Also in this case, annealing at 773 K leads to almost full intermixing between the Fe and Pt layers. Increase of the annealing temperature does not lead to significant changes in the composition distribution. Annealing of films with Ag intermediate layer causes again a moderate penetration of Ag into the FePt layer and segregation towards the free surface.

C. Pt/Fe and Pt/Ag/Fe films on Al$_2$O$_3$(0001)

Fig. 5 shows the 2D XRD images of Pt/Fe and Pt/Ag/Fe films sputter-deposited on Al$_2$O$_3$(0001) substrates after post-annealing at various temperatures. It is apparent that the (001) superstructure peak is present on the XRD images even after annealing at 773 K for both the Pt/Fe and Pt/Ag/Fe films. These peaks have low intensity as compared to the fundamental (111) reflection, but the intensity is not uniformly distributed along the diffraction ring and has a well defined maximum on the equatorial line, indicating the onset of (001)-texture formation. Additional XRD measurements showed the appearance of the low intensity (001) reflection for the Pt/Fe film after post-annealing at 623 K and for the Pt/Ag/Fe film after post-annealing at 673 K (Fig. 6). The intensity of the (001) reflection increases drastically with increasing annealing temperature, confirming the pronounced (001)-texture for samples post-annealing at 1073 K, which is slightly less pronounced in Pt/Ag/Fe films. However, in Pt/Ag/Fe, a strong (111) reflection remains.

Fig. 7 shows the corresponding SNMS concentration depth profiles of the Pt/Fe and Pt/Ag/Fe films after post-annealing at 773 K and 873 K. The results are very similar to those obtained for films deposited on MgO(001) and SrTiO$_3$(001) substrates. Even after annealing at 773 K, there is an almost homogeneous distribution of Pt and Fe in the Fe/Pt film. Increase of the annealing temperature does not lead to a substantial modification of the concentration profile. For the Pt/Ag/Fe film, full intermixing of the Fe and Pt layers with pronounced Ag surface segregation after post-annealing at 773 K was obtained.

The structural analysis of FePt films formed on Al$_2$O$_3$(0001) after post-annealing revealed $L_1_0$ ordering and a pronounced (001)-texture, thus a strong perpendicular magnetic anisotropy might be expected in these films. The magnetic properties of the annealed films were investigated by SQUID-VSM. M-H hysteresis loops were measured at room temperature in two geometries: magnetic field is applied in the film plane and out of the film plane. Figure 8 shows normalized M-H hysteresis loops obtained for Pt/Fe and Pt/Ag/Fe films after post-annealing at high temperatures.
The magnetization curves for all films are quite similar and a more or less isotropic behavior in magnetization reversal for the in-plane and out-of-plane field directions is observed. The coercivity of the Pt/Fe films after post-annealing at 973 K and 1073 K is 14.0 kOe and 15.8 kOe, respectively. For films with Ag intermediate layer, these values were increased up to 17.7 kOe and 24.2 kOe, respectively. This behavior can be explained by Ag diffusion to the grain boundaries, which results in exchange decoupling of FePt grains, which in turn enhances the coercivity. Furthermore, the still present (111) and (200) orientations are responsible for the isotropic magnetic properties due to the rather randomly oriented FePt grains.

XRD results presented above indicate that the deposition of Pt/Fe and Pt/Ag/Fe layered films onto single crystalline $\text{Al}_2\text{O}_3$(0001) substrates with hexagonal structure leads to decrease of the $L1_0$-FePt phase formation temperature as compared to MgO(001) and SrTiO$_3$(001) substrates with cubic lattice. Moreover, annealing of the films on $\text{Al}_2\text{O}_3$(0001) substrate results in pronounced (001)-texture formation. On the other hand, introduction of the Ag intermediate layer leads to the slight deterioration of the (001)-texture with the presence of a strong (111) reflection. Results of the SNMS depth profiling were very similar for films sputtered onto the all investigated single crystalline substrates; even after post-annealing at 773 K almost homogeneous intermixing of the Pt and Fe layers was observed. This fact indicates that there is no noticeable effect of the substrate type on the diffusion processes but its influence on the chemical ordering and texture formation is more significant.

The Ag intermediate layer increases the coercivity of the films after their post-annealing. This can be explained by decreasing the magnetic interaction between the $L1_0$-FePt grains due to their isolation. Isolated grains were formed because of the limited Ag solubility in FePt lattice and its grain boundary and surface segregation tendency (as was shown above). This conclusion is in agreement with the conclusions obtained in Ref. 26. Despite the pronounced (001)-texture, films deposited onto $\text{Al}_2\text{O}_3$(0001) substrates are magnetically isotropic, indicating the presence of chemically disordered A1 grains and $L1_0$ ordered grains with (111) and (200) orientations.

The observed differences in the texture formation in the films deposited onto different substrates can be explained by...
stresses, created by the mismatch between crystal lattices and by the difference of the thermal expansion coefficients of metallic layers and the substrate. These stresses are the origin of the strain favoring the growth of [001]-oriented grains.\textsuperscript{22} Stresses arising due to the difference in thermal expansion coefficients can be calculated from the equation\textsuperscript{27}

\[
\sigma = \Delta \alpha \Delta T E (1 - \mu),
\]

where \(\Delta \alpha\) is the difference in the thermal expansion coefficients between the substrate and the film, \(\Delta T\) is the temperature difference between room and the post-annealing temperatures, \(E\) is the elastic modulus of the film, and \(\mu\) is Poisson's ratio. As the elastic modulus is temperature dependent, and in our case the Fe layer interacts with the Pt, forming first A1-FePt and then L1\textsubscript{0}-FePt phases, we used the elastic modulus (180 GPa), Poisson’s ratio (0.33), and thermal expansion coefficient (10.5 \times 10^{-6} \text{ K}^{-1}) for bulk FePt for the estimation of the difference in the stresses occurring in the films deposited onto different substrates. Fig. 9 shows the calculated stresses that arise from thermal expansion mismatch as a function of temperature for the investigated samples. It is clear that the level of compressive stresses in the films deposited onto Al\textsubscript{2}O\textsubscript{3}(0001) substrates is much higher as compared to MgO(001) and SrTiO\textsubscript{3}(001) substrates.

FIG. 7. Composition profiles of films on Al\textsubscript{2}O\textsubscript{3}(0001) substrates after post-annealing at different temperatures: (a) Pt/Fe at 773 K; (b) Pt/Fe at 873 K; (c) Pt/Ag/Fe at 773 K; and (d) Pt/Ag/Fe at 873 K.

FIG. 8. SQUID-VSM M-H hysteresis loops of films annealed at different temperatures: (a) Pt/Fe at 973 K; (b) Pt/Fe at 1073 K; (c) Pt/Ag/Fe at 973 K; and (d) Pt/Ag/Fe at 1073 K.
These stresses promote the chemical ordering and texture formation in the films.

IV. CONCLUSION

In conclusion, sputtering of the Pt/Fe and Pt/Ag/Fe films onto Al$_2$O$_3$(0001) single crystalline substrates and following post-annealing leads to the formation of pronounced (001)-texture. Furthermore, the onset temperature for chemical ordering in these films is lower compared to films prepared on MgO(001) and SrTiO$_3$(001) substrates. It is important to note that there is no noticeable effect of the substrate choice on the diffusion process but its influence on the chemical ordering and texture formation is significant. Differences in the structural properties of the films deposited onto different substrates can be explained by the stress state that occurs during post-annealing. Also it was shown that introduction of the Ag intermediate layer is an effective method to increase the coercivity of the film.

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3T. Balat and D. Goll, "Large-area hard magnetic Li$_1$$_0$ FePt nanopatterns by nanoimprint lithography," Nanotechnology 22, 315301 (2011).