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Concurrent improvement of magneto-optical and perpendicular magnetic properties in CoPt/Ag stacked structures with ZnO intermediate thin layers

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The magneto-optical properties of stacked-layer structures consisting of hcp-$Co_{80}Pt_{20}$ thin films and noble metal underlayers were investigated under polar Kerr measurement conditions. For CoPt/Ag stacked films, insertion of a 2-nm-thick ZnO layer into the interface resulted in the concurrent improvement of the magneto-optical enhancement at the plasma edge of Ag and perpendicular magnetic anisotropy. An ideal square out-of-plane hysteresis loop with a large Kerr rotation of approximately 1.25° was obtained in the CoPt/ZnO/Ag stacked structure. Moreover, heat treatment in a vacuum resulted in further improvement of the perpendicular magneto-optical properties in the stacked structures with thin CoPt layers. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4793232]

The improvement of the magneto-optical (MO) Kerr and Faraday effects due to surface plasmon and localized plasmon resonances, magneto-photonic crystals, and diluted magnetic semiconductors has been studied extensively. A large MO effect is desirable in practical applications, such as MO recording media, MO spatial light modulators (SLMs), and magneto-plasmonic sensors. The effect of bulk plasmons on the MO properties in metals was initially investigated by Feil and Hass[1] and Katayama *et al.*,[2] and the enhancement of the Kerr rotation angle around the plasma (absorption) edge of noble metals was observed in layered-structures consisting of magnetic transition metals (MTMs) and noble metals (NMs). Co/Ag multilayers exhibit a relatively sharp MO peak at the plasma edge of Ag metal.[3] Strong MO enhancement due to surface plasmon resonance was also reported in Au/Co/Au multilayer structures under the total reflection condition,[4] and it has recently been reported that the MO effect is enhanced in nanostructures consisting of MTMs and NMs because of the localized plasmon resonance.[5–8] In addition to the large MO effect, perpendicular magnetic anisotropy is a prerequisite for applications such as high-density magnetic and MO recording media,[9] high-resolution MO-SLM,[10,11] and spin electronics devices.[12,13] Co–Pt alloys, multilayers, and nanostructures, which exhibit a relatively large MO effect at short wavelengths of blue light, are potentially useful for these applications because of their large perpendicular magnetic anisotropy and good chemical stability. For example, hcp-$Co_{80}Pt_{20}$-based films with a Ru underlayer are used as a high-density perpendicular magnetic recording medium in commercial products, and $L1_0$-$Co_{50}Pt_{50}$ composite films with Ag have been investigated as a next-generation high-density medium.[14,15] In next-generation magnetic recording systems, heated nanoscale spots based on near-field optics will be used as a new recording method.[16] Alternatively, a large MO effect has also been observed in diluted magnetic semiconductors of Co-doped ZnO.[17] The wide band-gap semiconductor ZnO, which has a small spin-orbit coupling implying a large spin coherence length, is expected to be applicable in spin electronics using magnetic tunnel junctions.[18,19] A large perpendicular magnetic anisotropy is also required in these applications because of the high density memory cells required in random access memories; thus Co–Pt based materials have been extensively investigated.

On the basis of the above discussion, it is clear that combinations of Co–Pt, Ag, Au, Ru, and ZnO have very interesting properties that are useful for many magnetic and MO applications. In the present study, the MO properties of hcp-$Co_{80}Pt_{20}$/NM (NM = Ag, Au, and Ru) stacked-layer structures with ZnO thin layers at the interface have been investigated under polar Kerr measurement conditions.

Co-rich CoPt thin films with 100-nm-thick Ag, Au, and Ru underlayers were deposited by magnetron sputtering on a glass substrate at ambient temperature. A 10-nm-thick ZnO seed layer with crystalline (001) orientation along the surface normal was deposited to improve the crystalline orientation of the fcc-Ag (111), fcc-Au (111), and hcp-Ru (001) underlayers. The ratio of the Co:Pt concentration, as measured by x-ray photoelectron spectroscopy, was approximately 80:20 at. %, and the structure of the stacked films was characterized by x-ray diffraction (XRD) measurements. The room-temperature polar Kerr rotation (θ_k) spectra in the range from 250 to 900 nm (1.38–4.96 eV) were measured at an applied field of 25 kOe.

Figure 1 shows the θ_k spectra and hysteresis loops for 5-nm-thick CoPt films directly deposited on the Ag, Au, and Ru underlayers (the hysteresis loops were measured at the photon energies shown by each arrow in the figure). The θ_k of the CoPt/Ag and CoPt/Au films was enhanced at the plasma edges of Ag and Au, respectively. Note that the Ag-underlayer sample exhibited a sharp θ_k peak at approximately 3.85 eV, which corresponded to the plasma edge of Ag, and the saturation θ_k was 1.03°, which was approximately 10 times that for the sample with the Ru-underlayer. The reflectivity (R) measured by using a spectrophotometer for the CoPt/Ag film markedly decreased to approximately 10% around the plasma edge of Ag; however, this sample exhibited a large figure of merit ($R\theta_k^2$) for MO applications,[9]

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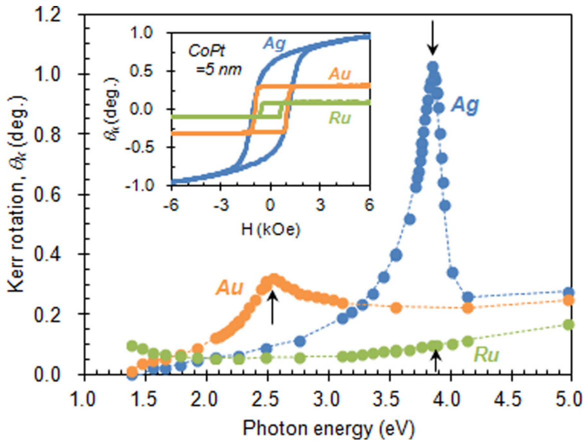

FIG. 1. Saturation Kerr rotation (θ_k) spectra measured at 25 kOe for CoPt films directly deposited on Ag, Au, and Ru underlayers. The inset shows the polar Kerr loops for the Ag, Au, and Ru-underlayer samples measured at the photon energies shown by each arrow.

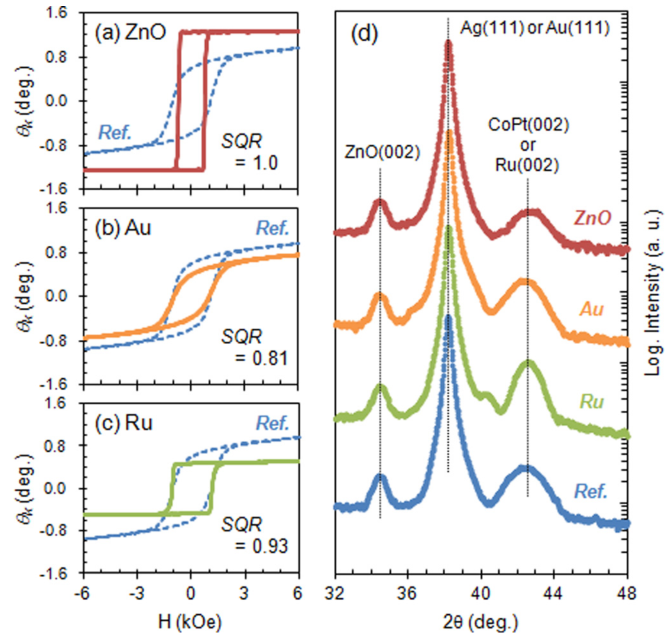

FIG. 2. Polar Kerr loops of CoPt/Ag stacked structures with 2-nm-thick (a) ZnO, (b) Au, and (c) Ru intermediate layers and without an intermediate layer (*Ref.*; dashed curves in each figure). An ideal square out-of-plane Kerr loop ($SQR = 1.0$) was obtained only in (a) the ZnO-sample. (d) XRD profiles for the CoPt/Ag stacked structures with and without the intermediate layers.

and it was approximately 3.8 times that for the Ru-underlayer sample. As can be seen in the hysteresis loops, a large θ_k was obtained for the Ag-underlayer sample, but the perpendicular squareness ratio (SQR) was less than unity. Comparing the Ag- and Au-underlayer samples, it can be seen that relatively good perpendicular magnetic properties were obtained only in the CoPt/Au stacked-film. However, the crystalline hcp-CoPt (001) orientation along the surface normal was observed in the XRD measurements for both samples, and the Ag and Au underlayers exhibited relatively good crystalline fcc-(111) orientation and nearly the same lattice distance. Therefore, it appears that the conditions of the CoPt initial growth layer contacting the underlayer are important for the development of perpendicular magnetic properties. In the CoPt films directly deposited on the Ag, Au, and Ru underlayers, both a large θ_k and an SQR of unity were not obtained. It is necessary to have both a large MO effect and a large perpendicular magnetic anisotropy for high-density MO recording and high-resolution MO-SLM.

Thus, several thin layers were inserted into the CoPt–Ag interface to investigate the influence of the interface conditions on the perpendicular magnetic properties. Figure 2 shows the hysteresis loops and θ–2θ XRD profiles for CoPt/Ag films with 2-nm-thick ZnO, Au, and Ru intermediate layers and without an intermediate layer (*Ref.*; dashed curves). It can be seen in Fig. 2(a) that the perpendicular magnetic properties of the CoPt film are markedly improved by inserting the ZnO intermediate layer, and an ideal square out-of-plane hysteresis loop ($SQR = 1.0$) with a large Kerr rotation angle of approximately 1.25° was obtained in the CoPt/ZnO/Ag stacked structure. The Ru intermediate layer also improved the perpendicular magnetic properties; however, the squareness ratio was less than unity ($SQR = 0.93$), and the θ_k value was markedly decreased to approximately half. ZnO was more effective to improve the perpendicular magnetic properties than Ru which has been usually used as an intermediate layer for high density perpendicular magnetic recording media. Conversely, the Au intermediate layer did not improve the perpendicular MO properties, although the CoPt film directly deposited on the Au underlayer exhibited relatively good perpendicular magnetic properties. For the CoPt/ZnO/Ag stacked-films, the XRD analysis suggests that the increase in the perpendicular magnetic anisotropy is not due to the improvement of the crystalline hcp-CoPt (001) orientation; it is likely attributed to the condition of the initial CoPt growth layer at the interface. In previous studies on Co-based multilayers, a large perpendicular magnetic anisotropy has not been reported in experimental and theoretical investigations for Co/Ag multilayers. Kingetsu and Sakai attributed the difference in perpendicular magnetic anisotropy between Co/Ag and Co/Au multilayers to the structural difference at the interface,[20] while Kyuno *et al.* reported that, on the basis of a first-principles calculation, the strength of the hybridization of the electronic states at the interface was important for the magnetic anisotropy of Co/NM (NM = Ag, Au, Cu, and Pt) multilayers.[21] In addition, it has recently been reported that the origin of the perpendicular magnetic anisotropy in Co/oxide stacked structures is attributed to the hybridization of the O and Co orbitals.[22,23] The results of the present study also suggest that the interface conditions are important for the development of perpendicular magnetic anisotropy in the CoPt/Ag stacked-systems. Further detailed experimental and theoretical investigations are required to clarify the origin of the perpendicular magnetic anisotropy. It is clear that the 2-nm-thick ZnO intermediate thin layer was remarkably effective at improving the perpendicular MO properties in the CoPt/Ag stacked structures. A large perpendicular magnetic anisotropy was previously reported for $L1_0$-$Co_{50}Pt_{50}$ films directly deposited on Ag underlayers;[24] however, $L1_0$-$Co_{50}Pt_{50}$ films generally require fabrication at high temperatures above 600 °C for $L1_0$-ordering. The hcp-$Co_{80}Pt_{20}$/ZnO/Ag stacked structures developed in this study, in contrast, are capable of making an ideal out-of-plane hysteresis loop upon fabrication at ambient temperature.

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Furthermore, it was found from the hysteresis loops shown in Fig. 2(a) that the MO effect was also enhanced by the ZnO intermediate layer. Figure 3(a) shows the influence of the ZnO intermediate layer thickness on the saturation θ_k spectra in the CoPt/ZnO/Ag stacked structures. An increase in θ_k and a peak shift to lower photon energy were observed with an increase in the thickness of the ZnO intermediate layer. All of the samples with ZnO intermediate layers exhibited a perpendicular *SQR* of approximately unity. For the stacked structures with a relatively thick ZnO layer of 14 nm in thickness, a new MO peak appeared near 3.3 eV in photon energy, which corresponds to the band-gap energy of ZnO. These peaks are attributed to an enhancement effect due to plasma (absorption) resonance of the ZnO/Ag underlayer. The complex Kerr rotation is related to the diagonal ($\varepsilon_{\mathrm{xx}}$) and off-diagonal ($\varepsilon_{\mathrm{xy}}$) elements of the dielectric tensor by the equation[25]

$$\theta_k + \mathrm{i}\eta_k \approx \frac{\varepsilon_{xy}}{\sqrt{\varepsilon_{xx}}(1-\varepsilon_{xx})}, \tag{1}$$

where η_k is the Kerr ellipticity. When the thickness (d) of magnetic layer formed on non-magnetic underlayer is thin enough as compared with the wavelength (λ) of incident light, the complex Kerr rotation is approximated by the following equation:[26]

$$\theta_k + \mathrm{i}\eta_k \approx \mathrm{i}\frac{4\pi d}{\lambda}\frac{\varepsilon_{xy}}{1-\varepsilon_{xx}^S}, \tag{2}$$

where ε_{xx}^S is the diagonal part of dielectric tensor of the non-magnetic underlayer. Therefore, the Kerr rotation is influenced by not only the dielectric tensor of magnetic layer but also that of non-magnetic underlayer. Near the plasma edge of Ag underlayer, a large Kerr rotation can be obtained because the real part of ε_{xx}^S approaches zero and the denominator in Eq. (2) has minimum.[1,3] Similarly, the enhancement of Kerr rotation around the band-gap energy of ZnO is calculated by using the optical constants[27] of ZnO. The origin of large Kerr rotation in the CoPt/ZnO/Ag stacked structure is attributed to the improvement of plasma enhancement effect due to the insertion of ZnO layer. Figures 3(b) and 3(c) show the reflectivity and figure of merit for these samples, respectively. The reflectivity decreased with increasing the thickness of ZnO intermediate layer, while the figure of merit increased by inserting ZnO layer. The stacked film with a 14-nm-thick ZnO layer exhibited the maximum figure of merit at the band-gap energy of ZnO, which was approximately 1.6 times that around the plasma edge of Ag for the sample without ZnO layer. Improvement in the perpendicular MO properties has also been achieved by adding an intermediate layer in transparent conductive Al-doped ZnO; therefore, the CoPt/ZnO/Ag stacked-system will be useful for magnetic and MO devices using spin injections.

The perpendicular MO properties in the CoPt/ZnO/Ag stacked structures were further improved by annealing in a vacuum. Figure 4 shows the MO properties measured at around the plasma edge of Ag for the as-deposited and annealed samples with various CoPt layer thicknesses. The ZnO intermediate layer thickness was 2 nm for all of the samples, and the annealing temperature and time were 400 °C and 30 min, respectively. The annealed samples were

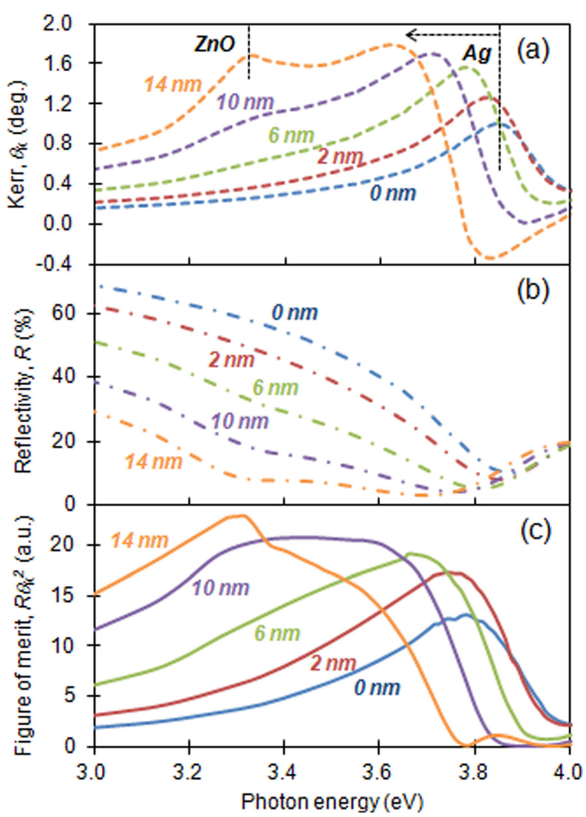

FIG. 3. Influence of the ZnO intermediate layer thickness on (a) the saturation Kerr rotation (θ_k), (b) the reflectivity (R), and (c) the figure of merit ($R\theta_k^2$) spectra for the CoPt/ZnO/Ag stacked films.

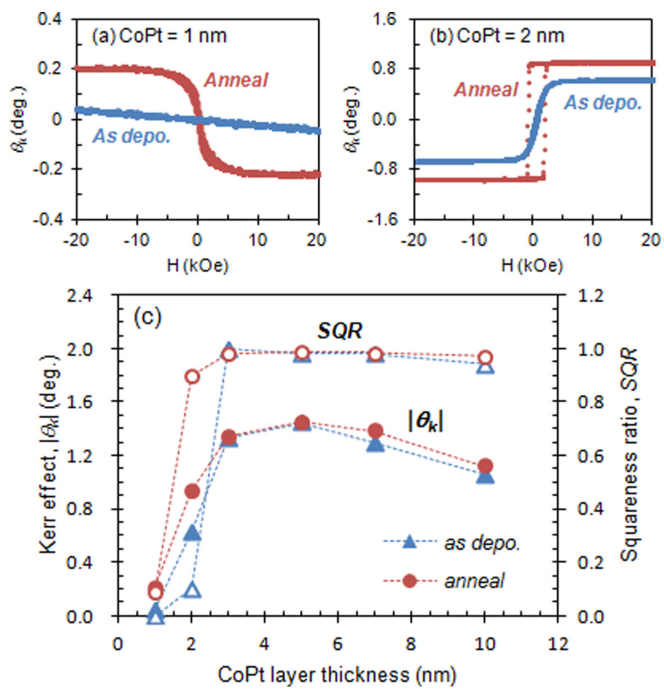

FIG. 4. Annealing effects on the perpendicular MO properties for CoPt/ZnO/Ag stacked films with various CoPt layer thicknesses. Polar Kerr loops of as-deposited and annealed samples with (a) 1-nm and (b) 2-nm-thick CoPt layers. (c) The CoPt layer thickness dependence on the absolute value of the polar Kerr rotation (closed symbol) and perpendicular squareness ratio (open symbol) for the as-deposited (triangles) and annealed (circles) samples.

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heated in the sputtering chamber at a base pressure of approximately 7×10^{-7} Pa immediately after deposition. Figure 4(c) shows the absolute value of Kerr rotation angle ($|\theta_k|$) at +25 kOe, and only a stacked film with 1-nm-thick CoPt layer exhibited a negative value. For the as-deposited and annealed samples, the $|\theta_k|$ increased with increasing CoPt thickness, and the maximum was obtained at a thickness of 5 nm. The $|\theta_k|$ peak position of the annealed samples shifted from 3.88 eV to 3.77 eV with increasing CoPt layer thickness. The appearance of a ferromagnetic hysteresis loop and improvement of the perpendicular MO properties were observed in the annealed samples with 1-nm and 2-nm-thick CoPt thin layers, respectively. These are probably attributed to the reason that the CoPt–ZnO interface becomes sharper by heat treatment, and it will be revealed by detail investigations of the microstructure of the interface. The square out-of-plane hysteresis loop and improvement of the perpendicular magnetic properties by heat treatment in the very thin CoPt films will be useful for magnetic tunnel junctions, because reduction of the magnetic layer thickness and annealing stability are required for these applications.

In conclusion, the MO properties of hcp-$Co_{80}Pt_{20}$ thin films deposited on Ag, Au, and Ru underlayers were investigated under polar Kerr measurement conditions. A large enhancement of the Kerr rotation angle was observed at the plasma edge of Ag in the CoPt/Ag stacked-films. The MO enhancement and perpendicular magnetic properties were concurrently improved by inserting a 2-nm-thick ZnO layer into the CoPt–Ag interface. An ideal out-of-plane hysteresis loop with a large Kerr rotation angle of approximately 1.25° was obtained in the CoPt/ZnO/Ag stacked structures. For the stacked structures with a relatively thick ZnO layer of 14 nm in thickness, a new MO peak appeared around the band-gap of ZnO, and the maximum figure of merit was obtained at this photon energy. The perpendicular MO properties of the stacked structures were further improved by annealing in a vacuum. The appearance of a ferromagnetic hysteresis loop and improvement of the perpendicular magnetic properties were also observed in the stacked structures with CoPt thin layers. An ideal square hysteresis loop with a large Kerr rotation in ultraviolet region will be especially useful for the high-density medium of next-generation MO recording systems with a short-wavelength laser.

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