Second-order spin-reorientation transition via magnetoelastic coupling in CoₚPd₁₋ₓ alloy films

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We report an observation of smooth spin-reorientation transitions from in-plane to normal-to-plane direction with increasing Pd overlayer thickness in CoₚPd₁₋ₓ alloy films, showing characteristics of a second-order transition (SOT). The magnetoelastic coupling term \( \Gamma \) plays a crucial role in such behavior via its contribution to the fourth- as well as second-order anisotropy constants. The nature of the SOT is characterized by \( -\Gamma \_1/\Gamma _2 = 0.12–0.47 \) determined for \( x = 0.25–0.40 \) in conjunction with lattice-misfit strains. The controlling spin orientation or anisotropy axis via magnetoelastic coupling without applying a magnetic field may be applicable to miniaturized memory and sensor devices.

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In this article, we investigate the SOT in CoₚPd₁₋ₓ alloy thin films with varying nonmagnetic Pd overlayer thickness \( t \_Pd \) as a driving parameter. By using relatively thick CoPd alloy layers of 30 Å, we can avoid many complex effects associated with the thickness variation in magnetic layer as well as many types of disorders at the interfaces. The CoₚPd₁₋ₓ alloy system is chosen for the following reasons. It was reported that the Co orbital moment is remarkably enhanced by Pd neighbors due to modified Co spin-polarized electronic structure. The orbital moment can play a crucial role in determining the spin orientation through spin-orbital coupling; thereby the strain evolution driven by the \( t \_Pd \) variation could result in the SRT. It was also reported that CoPd alloy films can have a perpendicular anisotropy. Here, we report clear evidence of the SOT and identify its mechanism by means of in situ magneto-optical Kerr effect (MOKE) magnetometry. It is found that the ratio of fourth- to second-order magnetoelastic coupling terms is a key factor governing not only the type of transition (SOT vs FOT) but also the nature of the SOT.

The spin-reorientation transition (SRT) in ultrathin magnetic films continues to grow in both fundamental and technological interests. This is because information storage technology is based on controlling the spin orientations. It is believed that the direction of easy magnetization in thickness- or temperature-driven SRT's is associated with the energy balance between surface (or interface) anisotropy preferring perpendicular magnetization and bulk dipolar (shape) anisotropy preferring an in-plane one.

Several groups have studied a variety of systems to better understand the novel transition behavior from in plane to normal to plane or vice versa. For example, a canting out-of-plane orientation of easy magnetization was observed in Co/Au (111) films using a scanning electron microscope with polarization analysis. Both abrupt and continuous SRT's were observed in a Ni/Cu (001) system by varying the Ni layer thickness. According to the transition type of spin reorientation, a smooth, continuous SRT through canted out-of-plane orientation and an abrupt, discontinuous one have been classified as a second-order transition (SOT) and a first-order transition (FOT), respectively. To explain both transitions, fourth-order \( K_4 \) as well as second-order \( K_2 \) anisotropy constants are introduced in a phenomenological equation of magnetic anisotropy energy. It is believed that the canting orientations, which are essential in the SOT, are caused by a nonzero higher-order anisotropy contribution.

Although higher-order anisotropy constants were intensively considered to understand thickness-driven SRT's in a Ni/Cu(001) system, the exact mechanisms of the different behaviors have not been clearly understood. It has also been reported that lattice-misfit strains induced by nonmagnetic or magnetic capping layers and morphology changes in an ultrathin regime promote SRT's, while just one monolayer (ML) thick capping layers remarkably increase perpendicular magnetic anisotropy through an interface anisotropy term. In these studies, complexities in the driving forces closely associated with the thickness variation seem to hinder a clear understanding of the mechanism of the SOT. Competing and/or cooperative effects on thickness-driven SRT's, such as a 1/\( t \) dependence due to interfaces (usually referred to as \( K_r/\Gamma_1 \)), lattice-misfit strains, and morphology changes in the ultrathin regime, make it difficult to identify the exact mechanism of the SOT.

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hysteresis loops with polar Kerr rotation loops shown here due to relatively small signals compared to the also observed in the longitudinal Kerr ellipticity loops ~ for particular, as for ~ for large , the SRT’s do not complete to the perpendicular for small , we used values of the magnetizations at an applied field maximum instead of . For those cases, we expect a larger error in the determination of magnetization components than for the loops fully saturated in a given field range.

In Fig. 3(a), we plot the easy magnetization orientations from the film normal at , defined as , vs for . For , the character of SRT with does not appear in vs . For a quantitative understanding of the continuous SRT with and its different behavior vs , spin orientations during the reversal were determined by a method utilizing not only but also . The vector analysis described elsewhere enables us to determine the magnetization vector components and normalized by the saturation magnetization , during the reversal for . For some cases where the loops are not saturated at all due to a field limit of in situ magnets, especially for thinner and larger , note in-plane and normal-to-plane directions, respectively. For all cases, we expect a larger error in the determination of magnetization orientations than for the loops fully saturated in a given field range.

The laser light of a wavelength of 632.8 nm was incident at 45° from the film plane.

Figure 1 shows the polar Kerr hysteresis loops, monitored by vs for each as noted. The initially slanted curves at each become squared with increasing except for and . Inverse trends with increasing were also observed in the longitudinal Kerr ellipticity loops (not shown here due to relatively small signals compared to the polar Kerr rotation loops). As shown in the evolution of the hysteresis loops with , the hard axis loops at become easy axis loops after a certain , also supported by the polar Kerr susceptibility defined by at , as shown in Fig. 2. The susceptibility plots clearly indicate characteristics of the SRT’s with such as the peaks and widths. This reveals that easy axis switches from the in-plane to the normal-to-plane direction occur through canted out-of-plane orientations, while the alloy layers with show in-plane magnetizations. In general, the SRT’s for small occur at thinner than for large with an exception for . The onset thickness of the transition from in-plane to perpendicular as well as the range of , for canted out-of-plane orientations, varies with . For and , the SRT’s do not complete to the perpendicular orientation as seen in the general trends of the loops. In particular, as for a fine feature of the loops at becomes a slanted square hysteresis. Whether it is real or not due to experimental artifacts or complexity originating from both an interfacial anisotropy term at the CoPd/Pd interfaces and the change in magnetostriction coefficient with , the character of SRT with does not appear in vs .

FIG. 2. Polar Kerr susceptibility vs for each vs . The peak positions and full widths at half maximum are indicated by the gray vertical and horizontal lines, respectively.
ruling out the determination of the magnetoelastic anisotropy constants only from canted angles.

To correlate $\theta_{H=0}$ obtained for canted out-of-plane orientations with $K_2$ and $K_4$, we use a phenomenological equation of the magnetic anisotropy energy $E = (K_2 + 2 \pi M_z^2) \cos^2 \theta + K_4 \cos^4 \theta$, where $\theta$ is the angle between the film normal and the magnetization direction. A positive sign of $K_2$, implies in-plane magnetization for convenience in this study while the positive sign convention is usually used for perpendicular in-plane magnetization for convenience in this study. Since the canted angles are caused by a nonzero $k_4$ contribution, we can determine $k_2$ and $k_4$ by fitting the experimental data set of $k_2/k_4$ vs $t_{pd}$ obtained from $\theta_{H=0}$ shown in Fig. 3(a) using a model to be proposed in the following. To elucidate the smooth SRT with $t_{pd}$ observed in this system, we assume that such behavior is caused mainly by strain evolution with $t_{pd}$ because only $t_{pd}$ is varied to result in the SRT for a given $x$ and because smooth transitions in a broad range of $t_{pd}$ ranging from 0 to 150 Å occur as shown in Figs. 1 and 2. If an interface anisotropy term at the Co/Pd/Pd interfaces were dominant in the observed SRT’s, a few ML Pd coverage would complete to switch to perpendicular orientations like a result reported by Engel et al. Therefore, the interface anisotropy term is not a dominating mechanism of the SRT’s at all. We believe that magnetoelastic anisotropy is dominating in relation to strain evolution with increasing $t_{pd}$ so that in the following model to explain the observed SRT’s we can assume that the magnetoelastic coupling term $\Gamma$ contributes to $k_{2(4)}$ in conjunction with a lattice-misfit strain $\varepsilon: k_{2(4)} = k_{c2(4)} + \Gamma_{2(4)} \varepsilon(t_{pd})$. The $k_{c2(4)}$ is the bulk magneto-crystalline anisotropy, assumed to be independent of $t_{pd}$, and $\varepsilon$ is a function of $t_{pd}$.

Numerical values of $\varepsilon$ vs $t_{pd}$ are required to determine $\Gamma_{2(4)}$ from data fits to the model. To obtain $\varepsilon$ vs $t_{pd}$ for a given $x$, we calculate the values of $\varepsilon$ at a coherent interface based on the experimental observations that the measured and calculated $\varepsilon$ in this system are somewhat in agreement with a factor of the magnitude. In addition, the trends of the SRT with $t_{pd}$ obviously show that the strain relaxation between the alloy layers and their overlayers is negligible up to $t_{pd} = 120$ Å. Thereby we can use the calculated coherent strains in the fits, as seen in the inset of Fig. 3(b). Strain values of the alloy layers increase with $t_{pd}$, depending on $x$ due to variations in the lattice parameters and biaxial modulus with $x$. For reliable fits, we made $k_{c2(4)}$ fixed by determining these values from other fits of the $M_z$ loops with $t_{pd} = 0$ with $h = 2k_2M_z + 4k_4M_z^2$, where $h = H/M_z$. The loops for $t_{pd} = 0$ have no strain effects caused by the Pd overlayers, i.e., $\varepsilon = 0$ at $t_{pd} = 0$ in the model. Resulting fits with a constraint of $20° \leq \theta \leq 70°$ are shown in Fig. 3(b), in good agreement with the measured data. The constraint is for better fits since the solution of $\cos^2 \theta = -k_2/k_4$ is reliable only for the case of canted orientations.

In Fig. 4(a), we plot the values of $\Gamma_{2}$, $\Gamma_{4}$, and the ratio of $-\Gamma_{2}/\Gamma_{4}$ determined from the fitting process. The opposite sign of $\Gamma_{2}$ and $\Gamma_{4}$ reveals their opposite character in the SRT: a negative value of $\Gamma_{2}$ prefers perpendicular magnetization (according to the definition in this study) while a positive one of $\Gamma_{4}$ prefers in-plane magnetization. First, we stress a clear peak in $\Gamma_{4}$ at $x = 0.35$ while the magnitude of $\Gamma_{2}$ monotonically decreases with $x$. The different values of $\Gamma_{2(4)}$ between given $x$’s explain the different behaviors of the evolution of the $\theta^K_{\theta}$ loops shown in Fig. 1. The values of $k_2$ and $k_4$ vs $t_{pd}$ seen in Fig. 4(b) are determined from both the fitting values of $\Gamma_{2(4)}$ and the calculated $\varepsilon$’s shown in the inset of Fig. 3(b). The values of $k_2$ generally decrease rapidly.
from a positive at a thinner $t_{pd}$, cross a zero value, and then slowly increase in a negative sense in a wide range of thicker $t_{pd}$.

An anisotropy-constant phase diagram is very useful to better understand the SRT with varying parameters.\textsuperscript{7} Our result of the $\bar{k}_2-k_4$ anisotropy flows vs $t_{pd}$ is displayed in Fig. 5. Initial spin states with $t_{pd}=0$ Å for each $x$ are marked within a small box. Different phases marked by the thick gray lines clearly show the onset of $t_{pd}$ between in-plane, canted and perpendicular, depending on the relative strength of $\Gamma_2$ and $\Gamma_4$. The onset for reorientation from in plane to canted out of plane ($1 \rightarrow 2$) is determined by $\bar{k}_2=0$ while the onset between canted and perpendicular ($2 \rightarrow 3$) is governed by $k_4/\bar{k}_2=-1/2$.\textsuperscript{9} The corresponding onset of $t_{pd}$ is summarized in Fig. 5. The large ratios of $-\Gamma_4/\Gamma_2=0.12 \pm 0.03-0.47 \pm 0.12$ obtained for $x=0.25-0.40$ result in canted out-of-plane magnetization over a wide range of $t_{pd}$, since $\Gamma_4$ works against $\Gamma_2$, preferring perpendicular magnetization as tensile strains evolve with $t_{pd}$. It is also clearly seen in Fig. 5 that the slopes of the $t_{pd}$-driven SRT’s in the $\bar{k}_2-k_4$ plane are equal to the values of $\Gamma_4/\Gamma_2$ for each $x$. Thus the ratio of magnetoelastic coupling terms, $\Gamma_4/\Gamma_2$, determines the characteristics of the SRT in conjunction with lattice-misfit strains as well as the nature of the SOT, whether it is first or second order.

To speculate on the microscopic origin of the observed SRT behavior, we consider that the spin-polarized electronic structure of Co modified by Pd neighbors strongly enhances the Co orbital moment at their interfaces as reported by Kim and Kortright.\textsuperscript{20} Hybridization of interfacial Co and Pd atoms depending on $x$ would make a modification of Co spin-orbital coupling $\xi$ associated closely with $\Gamma$ since Pd has $\xi$ being three times larger than Co.\textsuperscript{29} However, it is still unknown why an alloy film with $x=0.35$ yields the largest value of $-\Gamma_4/\Gamma_2=0.47 \pm 0.12$ among given $x$’s. The possible origins might be a relative atomic configuration of Co and Pd atoms and/or a spin-polarized electronic structure varying with $x$.

In summary, we have observed second-order spin-reorientation transitions in Co$_{1-x}$Pd$_x$ alloy films through magnetoelastic coupling in conjunction with lattice-misfit strains changing with Pd overlayer thickness. The ratio of magnetoelastic coupling terms, $\Gamma_4/\Gamma_2$, determines the characteristics of the SRT as well as the nature of the SOT, whether it is first or second order. It is worthwhile to address the fact that controlling the spin orientation or anisotropy axis via strain modulation without applying an external magnetic field may be of technological importance in miniaturized magnetic memory and sensor devices as a potential switching mechanism.

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A simplified analytic formula of $p$- and $s$-polarization MOKE signals for an arbitrary magnetization direction is derived as follows [C.-Y. You and S.-C. Shin, Appl. Phys. Lett. 69, 1315 (1996)]: $\theta + i \eta = (\theta_s^L, \eta_s^L) + i(\eta_y^L, \eta_z^L)$, where $\theta$ and $\eta$ are the measured Kerr rotation and ellipticity, respectively, of both $p$- and $s$-polarizations. $m_{y,z}(L)$ is the direction cosine. $\theta_s^L$ and $\eta_s^L$ are the corresponding values obtained at the saturated (maximum here) field applied along in-plane ($y$) and normal-to-plane ($z$) directions. Since there are distinct sensitivities of both $\theta$ and $\eta$ to the $y$ and $z$ components of the magnetization vector, the derived equation can be also used to determine the magnetization components during the reversals by using the simultaneously measured Kerr rotation and ellipticity of only a $p$-polarization. Details about the vectorial analysis will be reported elsewhere [J.-W. Lee, J. Kim, S.-K. Kim, J.-R. Jeong, and S.-C. Shin (unpublished)].

The magnetostriction coefficient $\nu$ vs $t_{Pd}$, measured in situ using an optical displacement detecting method, was found to be almost constant over all the $t_{Pd}$ within experimental errors, which supports this model. Our in situ stress measurements during the deposition of Pd overlayers indeed displayed a tensile stress evolution with $t_{Pd}$.

The elastic strain energy density per unit area for a bilayer, having comparable thickness of individual layers, can be written as $E_{el} = e_A^2 Y_A + e_B^2 Y_B$, where $e_A(B)$, $Y_A(B)$, and $t_A(B)$ are the coherent strain, the biaxial elastic modulus for a (111) plane, and the thickness of the $A(B)$ layer, respectively. Together with the boundary condition $a_A(1+e_A)\approx a_B(1+e_B)$ at a coherent interface in continuum elastic theory, the minimization of $E_{el}$ with respect to $e$ yields an equilibrium strain $e_A^B = [(a_B - a_A) a_A t_A Y_B]/[a_A^2 t_A Y_A + a_B^2 t_B Y_B]$, where $a$ is the interatomic distance in a given plane.

Here the $h-M_z$ relationship was derived in a similar way described in the text by minimizing the energy involving the Zeeman energy term of $-HM_z \cos \theta$ with respect to $\theta$. A. S. Chakravarty, *Introduction to the Magnetic Properties of Solids* (Wiley, New York, 1980), p. 66.