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Spin-orbital coupling induced four-fold anisotropy distribution during spin reorientation in ultrathin Co/Pt multilayers

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In this work, we synthesized (Co(t)/Pt)3 multilayers and quantitatively determined thickness and temperature dependence of spin reorientation transition (SRT) and perpendicular magnetic anisotropy (PMA) using ferromagnetic resonance measurement. The critical thickness for PMA switching as well as tremendous magnetic anisotropy change up to 645 Oe once the temperature varies from 25 °C to −153 °C are demonstrated. More interestingly, a four-fold symmetry of magnetic anisotropy was found to be prominent during the SRT. By conducting magnetic simulation with involving high order energy term, we highly related this phenomenon to the strong spin-orbital coupling, which is considered to be the major energy term to tip the balance between the surface anisotropy and shape anisotropy. These results provide an opportunity for better understanding the transition behaviors which is essential for PMA structure preparation and their related devices. Published by AIP Publishing. [http://dx.doi.org/10.1063/1.4973884]

Since IBM announced the first hard disk drive in 1956, the recording densities keep increasing dramatically with a sharp scaled down track pitch and bit-cell length. This requires for size decreasing films,1,2 which will meet their collapse when the grain sizes reach superparamagnetic or thermal decay limits.1 As an alternative, the perpendicular magnetic anisotropy (PMA) materials, such as Co/Pt3 and Cu/Ni,4 shade into hot spots because of their high density and thermostability.5,6 They can often be realized by means of spin reorientation transition (SRT) in ultrathin magnetic films driven by the variation of thickness and temperature, exhibiting a promising way that enables ultrahigh density information storages.7 Experiments have demonstrated that magnetic easy axis can rotate between out-of-plane and in-plane directions in ultrathin magnetic films as film thickness or temperature changes—SRT.8,9 In these films, symmetry at the film-substrate interface is broken and gives rise to SRT through exchange coupling.8 However, the mechanism and process of SRT with critical structures have not received consensus.10 The spin-orbit operator, which is important in lower symmetries of ultrathin magnetic films, is usually ignored during the SRT to reduce the fundamental computational difficulty of this integration.11,12 Thus, further investigations on SRT are required from both practical and fundamental point of view, which is crucial for device applications. To quantitatively analyze SRT, discussion on energy distribution is needed. Kisielewski reported that the energy evolution during this transition can be described with anisotropy constants.13 Researchers have demonstrated that volume contribution KV and surface contribution KS have a combined action on the effective magnetic anisotropy energy (MAE) K1, and the relation can be approximately expressed by8,10,14

\[ K_1 = K_V + 2K_S/t_{Co}, \]

where t_{Co} is the thickness of Co single layer. Negative surface anisotropy KS causes reversed thickness dependence in the Co ultrathin multilayers. It is found that the decreasing thickness favors in-plane magnetization for Ni (Ref. 4) and out-of-plane magnetization for Fe (Refs. 15 and 16) and Co.4,13 In the Co ultrathin multilayers, it is because that the surface magnetic anisotropy (SMA) increases as the film thickness and temperature decreases and eventually will be strong enough to overcome demagnetizing field, aligning the magnetization to the perpendicular plane.17 Given the weak magnetic signals of ultrathin films, only magneto-optics and ferromagnetic resonance (FMR) methods are utilized for SRT investigation.14,18,19 Moreover, during critical SRT vicinity, the magnetic easy axis lies in intermediate state which is far from both in-plane and out-of-plane direction—the so called four-fold anisotropy distribution. Studies on this four-fold symmetry still remain theoretically and technically challenging. Until now, there are different opinions about the mechanism and the process of SRT in four-fold structure,10 some reports considered that SRT proceeds through a canting state,20 while others claim a coexistence of both “in-plane” and “out-of-plane” magnetization states.10,21 Moreover, reversible SRT has been observed as a function of temperature.22 Nevertheless near the transition point,
whether the magnetic properties are determined by thermal equilibrium or relaxation phenomena is still not clear.\textsuperscript{15} Besides the thermal stability of potential applications, temperature dependence is also of fundamental importance since all theoretical descriptions are based on a thermodynamic approach.\textsuperscript{15,23}

Here, we fabricate a PMA structures of Si(100)/Ta(3 nm)/Pt(1 nm)/(Co(t\textsubscript{Co})/Pt(1 nm))\textsubscript{3}/Ta(2 nm) with t\textsubscript{Co} = 0.3, 0.5, 0.7, 0.8, 0.9, 1.0, and 1.1 nm by magnetron sputtering. The base pressure of 2 × 10\textsuperscript{−7} Torr increased to 3 mTorr during evaporation. Both magneto-optics methods were utilized to study the SRT behaviors of (Co/Pt)\textsubscript{3} multilayers with varied thicknesses and temperatures. For the typical four-fold sample at the SRT, a 401 Oe upward shift for in-plane resonance field (H\textsubscript{r}) and a 645 Oe downward shift for out-of-plane were demonstrated. What is more, high order energy terms in the magnetic anisotropy were calculated under various conditions. We achieved well-agreement between the experimental FMR results at thickness/temperature SRT vicinity and their theoretical fittings. In our theoretical analysis, up to sixth-order energy terms were taken into account, which appears to be essential for the explication of the magnetic behavior at critical transition area. Our work provides a way to adjust the direction of magnetization easy axis and also holds potential applications in high-density perpendicular magnetic recording.

Figure 1(a) shows the out-of-plane hysteresis loops for 0.3, 0.5, 0.7, and 0.9 nm (Co/Pt)\textsubscript{3} films measured by the MOKE system at room temperature. Faraday rotation and polar Kerr effect were measured as a function of perpendicular applied magnetic field. It is found that all the four ultrathin films have well-established square shapes, indicating a perpendicular easy axis. One can see that the thinner the thickness the easier the perpendicular direction. However, if the film is too thin, the coercive field will drop down to 100 Oe, which makes it hard to maintain the stability of magnetization state. Angular dependence of FMR for different samples were studied at room temperature as shown in Fig. 1(b). It was carried out in resonant cavity and measured by the rotation of external magnetic field which was applied in the normal plane. For all the polar diagram, 0° represents in-plane direction while 90° means out-of-plane direction.

Larger resonance field indicates a higher magnetic energy, corresponding to the hard magnetization direction. Therefore, upon the thickness decreases, the magnetic easy axis is transforming from the in-plane (blue loop) to the out-of-plane direction (i.e., PMA, (white loop)).

Figure 2 displays magnetic domain evolution and dynamic magnetization reversal process for 0.5 nm (Co/Pt)\textsubscript{3} film. In the beginning, this film was completely polarized with a large perpendicular magnetic field so that all the magnetic moments pointed to the downward (dark region). Then, an opposite perpendicular magnetic field was gradually applied until it passes the coercive field. During this process, bubble domain structures with 180° domain wall appear, and by subtracting the initial background image, contrasts can be obtained as shown in Figure 2 (Multimedia view).

Despite the high sensitivity and good spatial resolution the magneto optics may have, it is limited in analyzing the spatial distribution of anisotropy energies. Thus, we studied temperature and angular dependence of H\textsubscript{r} for five different Co thicknesses through FMR measurements. As shown in Fig. 3(a), the external magnetic field was applied in the x-y plane. The angular dependence was measured by the rotation of 0\textsubscript{M} while the directions of magnetization M was illustrated by 0\textsubscript{M} and 0\textsubscript{M}. The results show that the magnetic easy axis transforms from the in-plane to the out-of-plane direction when t\textsubscript{Co} decreases from 1.1 nm to 0.7 nm as shown in Figs. 3(b)–3(f). For each thickness, lowering temperature makes the magnetization process harder (easier) along the in-plane (out-of-plane) direction. For both thickness and temperature driven SRT, the magnetic easy axis starts to rotate away from the in-plane direction and results in a four-fold symmetry of magnetic anisotropy at the critical vicinity of the spin reorientation, indicating a strong competition between surface anisotropy and demagnetization. In fact, the surface anisotropy can overcome shape anisotropy 4πM\textsubscript{S} in ultrathin Co/Pt multilayers, yielding the magnetization perpendicular to the films.\textsuperscript{4,12,24} Our experimental results go well with early theoretical studies.\textsuperscript{4,12,24} Typical transition state with four-fold shapes of magnetic anisotropy transition can be seen in Fig. 3(c), the sample with critical thickness (1 nm). When temperature drops from 25°C to −153°C, the

FIG. 1. Illustration of PMA and SRT with experimental results measured at room temperature. (a) Hysteresis loop for films with PMA measured with MOKE system. (b) Angular dependence of FMR for thicknesses near transition area. For all the polar diagram, 0° represents in-plane direction while 90° means out-of-plane direction.
in-plane $H_i$ of 1 nm sample obtains a 401 Oe upward shift while the out-of-plane $H_i$ possesses a remarkable 645 Oe downward shift.

To further reveal the internal physics underlying the transition, magnetic simulation involving high order energy terms was carried out in this paper. In order to precisely interpret magnetic behaviors for the transition area, up to sixth-order terms have been included in the energy density function for the hexagonal system as expressed in the following: \[ E = -HM \sin \theta_M \cos (\phi_H - \phi_M) + \frac{1}{2} 4\pi M^2 \sin^2 \theta_M \sin^2 \phi_M - (K_1 + 2K_2 + 3K_3 (\cos^4 \theta_M - 2 \cos^2 \theta_M)) \sin^2 \theta_M \sin^2 \phi_M + (K_2 - 3K_3 \cos^2 \theta_M) \sin^4 \theta_M \sin^4 \phi_M - K_3 \cos^6 \theta_M, \] (2)

where the first term is Zeeman energy, the second is the magnetostatic energy, and the remaining terms are the anisotropy energies with the $c$ axis parallel to the $y$ axis. \(^{19}\) The definition of angles is the same with the measurement configuration mentioned in Fig. 3(a). $K_1$, $K_2$, and $K_3$ represent the second-, fourth-, and sixth-order anisotropy constants, accordingly. The effective values of $K_1$, $K_2$, and $K_3$ in Co films depend on the volume magnetocrystalline anisotropy, the SMA, and the stress anisotropy. \(^{19}\) We calculated $K_{1,eff}$ according to the resonance field obtained by FMR. Since the dc field $H$ is applied in the normal plane, the magnetization $M$ balances at $\theta_M = \pi/2$. The $M$ free precession equation can be given by

\[
\frac{\partial \theta}{\partial \gamma} = \frac{-1}{M^2 \sin^2 \theta_M} \left[ \frac{\partial^2 E}{\partial \theta_M^2} \frac{\partial E}{\partial \phi_M^2} - \left( \frac{\partial^2 E}{\partial \theta_M \partial \phi_M} \right) \right],
\]

(3)

where the definition of angles is also the same with what we mentioned above. Neglecting magnetic elastic effect and interlayer exchange coupling, the ferromagnetic resonance equation (4) can be derived from energy density function Eq. (2) and $M$ free precession equation (3)

\[
\frac{\partial \phi}{\partial \gamma} = \frac{1}{H \cos (\phi_H - \phi_M) + (4\pi M - H_{A1} - H_{A2}) \cos 2\phi_M} + H_{A3} (3 \sin^2 \phi_M \cos^2 \phi_M - \sin^4 \phi_M) \times H \cos (\phi_H - \phi_M) - (4\pi M - H_{A1} - H_{A2} - 2H_{A3}) \times \sin^2 \phi_M - (H_{A2} + H_{A3}) \sin^4 \phi_M, \]

(4)

where $\omega$ is angular frequency for resonance (9.3 GHz $\times 2\pi$), $\gamma$ is the gyromagnetic ratio of 2.8 MHz/Oe, $H$ is the resonance field, $4\pi M$ is 17.7 kOe at room temperature and 18 kOe at low temperature, $H_{A1} = 2K_1/M$, $H_{A2} = 4K_2/M$, and $H_{A3} = 6K_3/M$. Three anisotropy fields can be calculated by the measured $H_i$ and they correspond to the second-, fourth-, and sixth-order energy terms, respectively. The effective magnetic anisotropy constant $K_{1,eff}$ can be given by

\[ K_{1,eff} = K_1 - 2\pi M^2 \sin^2 \theta_M. \]

(5)

where $K_1$ can be expressed with Eq. (1). \(^{10,14}\)

The determination of $K_v$ and $K_p$ presented in Eq. (1) can be obtained by plotting surface magnetic energy product $K_{1,eff} \times t_{Co}$ versus $t_{Co}$ for Co/Pt multilayers, as demonstrated in Fig. 4. A positive $K_{1,eff}$ corresponds to a magnetization favoring perpendicular direction. \(^{25}\) The negative slope, refers to a negative volume anisotropy $K_v$, describes the case of a preferred direction parallel to the layer plane. \(^{25}\) Intercept at $t_{Co} = 0$ indicates a positive interface anisotropy $K_p$ with the perpendicular magnetization. \(^{25}\) When $t_{Co}$ is less than a critical thickness $t_{c}$, $-2K_v/K_p$, in this case $\sim 10$ Å, SMA surpasses volume effect, rotating magnetization from the in-plane to perpendicular direction. Volume energy is the source of demagnetizing field which is usually responsible for in-plane magnetization, and can be overcome in ultrathin films. \(^{25}\) Besides, our computing results also reveal a temperature effect. When the temperature decreases from 25 $^\circ$C to $-153$ $^\circ$C, $K_{1,eff}$ shifts upward, leading $t_{Co}$ varies from 9.4 to 10 Å. The results imply that the easy axis generally rotates to perpendicular direction and a larger transition thickness is...
required for a comparable demagnetizing energy to override
the enhanced surface energy.

Theoretical fitting process containing high-order energy
terms was carried out for theoretical interpretation. Fig. 5(a)
shows both experimental (symbols + lines) and simulative
(green dashed lines) angle dependence of FMR field for the
thickest and thinnest films studied at room temperature. 1 nm
Co layer was also investigated for its strong temperature var-
ciation and corresponds to four-fold anisotropy distribution,
which are demonstrated in Fig. 5(b). The fitting calculation
was based on Eq. (4). The high-order (fourth and sixth)
energy terms relate to the spin-orbit interaction, which
mainly influences the fitting veracity at intermediate angles.
The spin-orbit interaction is associated with the broken sym-
metry and the smaller coordination number of atoms at the
interface/surface.26 Magnetic anisotropy energy (MAE) of
Co films can be calculated as a function of spin quantization
direction and smallest in the hard direction and it will
increase for lower symmetries.12 Therefore, a large MAE
can be expected in ultrathin films due to the reduced symme-
try of distorted lattices, interfaces, surfaces, and the presence
of microscopic roughness.12 Neel’s pair model has proposed
that a roughness enhancement by four orders of magnitude
in ultrathin films is expected compared with that of bulk
materials.12,27 As for our Co ultrathin films, the interface/sur-
face effects predominate, and the hexagonal symmetry is
lower than that of cube. Therefore, a fourth-order anisotropy
constant is necessary for the energy calculation (Fig. 5(a)).
However, if the thickness and temperature are close to the
critical vicinity of the spin reorientation, fourth order cannot
guarantee an accurate calculation. That is why we involved
the sixth-order energy term to assure a well-fitted data (Fig.
5(b)), which indicates a strong spin-orbital coupling taking
place in lower symmetries of ultrathin magnetic films.

Based on Eqs. (1), (4), and (5), energy terms are calcu-
lated for all the samples ranging from 7 Å to 11 Å. The angle
dependences of FMR field at 25°C and −153°C were fitted
involving all the high-order energy terms. The results of the fit-
ting procedure are reported in Table I. During the given ranges
of three anisotropy fields, well-fitted FMR versus angle curves
were obtained. H1 (corresponding to K1,eff shown in Fig. 4)
shows an upward trends when the temperature decreases. H3
is not that sensitive and hold a relatively wide change interval
but it cannot be ignored when considering four-fold shapes
(room temperature 8, 9 Å, all temperature of 10 Å, and low
temperature 11 Å, as shown in Fig. 3). The well-fitted data
shown in Fig. 5(b) can never be obtained when H3 = 0.

According to Eq. (4), anisotropy fields phase diagram
are also constructed to study the influence of high energy
terms’ influence on easy axis. The H3/H1 phase diagrams
for different values of H4 are shown in Fig. 6. One can
identify three stability regions: in-plane, intermediate, and
perpendicular. For H4 > 1.93 × 104 Oe, no SRT is possible,
and if H4 decreases, the easy axis gradually rotates to tilted
orientation when H4 varies between −600 and 2000 Oe.
When H3 changes from −500 Oe to 0 Oe and back to
1000 Oe, the intermediate region is expanding, conforming
the necessity for the transitional region (Multimedia view).

In conclusion, we have investigated the thickness and
temperature driven SRT in ultrathin (Co/Pt)3 films. For both
types of SRT, the rotation of magnetic easy axis can give
rise to a four-fold anisotropy distribution. We find the exis-
tence of a thickness transition point as magnetic easy axis
transforms from in-plane to out-of-plane direction. This transi-
tion Co thickness goes down as the temperature decreases.
Meanwhile, the sixth-order energy term has to be involved to

FIG. 4. Thickness dependence of K1,eff tCo calculated by ferromagnetic
resonance equation (Eq. (4)) for room and low temperature. Lines are linear
fitting results for the calculation.

FIG. 5. Typical experimental and fit-
ting results. (a) Curves for completely
in-plane (red, 1.1 nm) and out-of-plane
(blue, 0.7 nm) samples at room temper-
ature. (b) Curves for critical transition
sample (1 nm) at room (red) and low
(low) temperature.
TABLE I. Values of $H_A1$, $H_A2$, and $H_A3$ for all SRT samples while at room and low temperature. This calculation is based on ferromagnetic resonance equation (Eq. (4)). Results for four-fold samples is in bold.

<table>
<thead>
<tr>
<th>$t_{CO}$ (Å)</th>
<th>$H_{SRT}$ (Oe)</th>
<th>$H_{LT}$ (Oe)</th>
<th>$H_{SRT}$ (Oe)</th>
<th>$H_{LT}$ (Oe)</th>
<th>$H_{SRT}$ (Oe)</th>
<th>$H_{LT}$ (Oe)</th>
</tr>
</thead>
<tbody>
<tr>
<td>7</td>
<td>[20 220 – 147, 20 220 + 153]</td>
<td>20 937 ± 200</td>
<td>527 ± 177</td>
<td>[763 – 213, 763 + 137]</td>
<td>0 ± 500</td>
<td>[0 – 300, 0 + 40]</td>
</tr>
<tr>
<td>8</td>
<td>[19 100 – 150, 19 100 + 200]</td>
<td>20 605 ± 200</td>
<td>500 ± 100</td>
<td>450 ± 200</td>
<td>[600 – 350, 600 + 400]</td>
<td>[600 – 600, 600 + 400]</td>
</tr>
<tr>
<td>9</td>
<td>18 080 ± 100</td>
<td>[19 250 – 150, 19 250 + 100]</td>
<td>[620 – 100, 620 + 60]</td>
<td>500 ± 100</td>
<td>[460 – 180, 460 + 140]</td>
<td>0 ± 300</td>
</tr>
<tr>
<td>10</td>
<td>17 365 ± 65</td>
<td>18 500 ± 200</td>
<td>700 ± 100</td>
<td>700 ± 70</td>
<td>450 ± 150</td>
<td>300 ± 200</td>
</tr>
<tr>
<td>11</td>
<td>[15 896 – 370, 15 896 + 230]</td>
<td>[16 400 – 100, 16 400 + 200]</td>
<td>[574 – 224, 574 + 326]</td>
<td>800 ± 150</td>
<td>0 ± 500</td>
<td>[800 – 200, 800 + 100]</td>
</tr>
</tbody>
</table>

FIG. 6. Evolution for the direction of magnetic easy axis. The blue and red area means completely in-plane and out-of-plane direction while gradient colors represent intermediate region. $H_A3$ increases from $-500$ (a) to $0$ (b) and eventually to $1000$ Oe (c), and the intermediate region is expanding. (Multimedia view)[URL: http://dx.doi.org/10.1063/1.4973884.2]

guarantee an accurate interpretation of the magnetic behavior at the critical transition point. The high order anisotropy constant is a strong indication of the non-negligible spin-orbital coupling taking place in this lower symmetry systems.

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2Wikipedia, 2016-6-27.