Determination of perpendicular magnetic anisotropy in ultrathin ferromagnetic films by extraordinary Hall voltage measurement

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A magnetometric technique for detecting the magnetic anisotropy field of ferromagnetic films is described. The technique is based on the extraordinary Hall voltage measurement with rotating the film under an external magnetic field. By analyzing the angle-dependent Hall voltage based on the Stoner–Wohlfarth theory, the magnetic anisotropy field is uniquely determined. The present technique is pertinent especially for ultrathin films with strong intrinsic signal, in contrast to the conventional magnetometric techniques of which the signal is in proportion to the sample volume and geometry. © 2009 American Institute of Physics. [doi:10.1063/1.3262635]

Thin ferromagnetic films, which have perpendicular magnetic anisotropy, provide perspective challenges in high density data storage applications such as the hard disk,1,2 the spin-transfer-torque random access memory,3 the racetrack memory,4 etc. The static and dynamic properties are essentially influenced by the strength of the anisotropy in these materials, and thus, precise measurement of the anisotropy is of importance in designing and optimizing the device performance. However, since most of the films with the perpendicular magnetic anisotropy are realized in ultrathin layered structures typically with a-few-angstrom thickness,5–7 it is hard to detect the weak signal from the small volume by use of the conventional bulk techniques such as the torque magnetometry and the ferromagnetic resonance. In addition, the films with the perpendicular magnetic anisotropy generally exhibit complex magnetization reversal behaviors in competition between the wall motion and the nucleation other than the coherent rotation.8–10 It is thus hard to extract the sole information of the anisotropy from the complex magnetization process.

In this paper, we propose a simple experimental technique to measure the perpendicular magnetic anisotropy field. The technique is based on the extraordinary Hall effect measurement, which preserves strong signal even in ultrathin films; thus the effect is used as a useful tool for measuring the magnetization state.11 The extraordinary Hall voltage is measured with rotating an external magnetic field in a small angle around the easy axis and then analyzed based on the Stoner–Wohlfarth theory with small angle deviation to extract the value of the anisotropy field. We confirm the perfect consistency between the experimental data and the theoretical prediction, which evidence the validity of the technique and consequently provide the precise experimental value of the anisotropy field.

For a magnetic system with uniaxial magnetic anisotropy, the magnetic energy $E$ of the system is given by $E=-K_U \cos^2 \theta - M_S H \cos(\theta - \phi)$, as first proposed by Stoner and Wohlfarth.12 Here, $K_U$ is the uniaxial anisotropy, $M_S$ is the saturation magnetization, and $H$ is the external field. The angles $\theta$ and $\phi$ are the angles of the magnetization and the external field from the easy axis, respectively. Note that the easy axis is normal to the film for the case of the perpendicular magnetic anisotropy. Normalized by $K_U$, the equation is rewritten as $E=E/K_U=K_U - \cos^2 \theta - 2\alpha \cos(\theta - \phi)$ with a single characteristic parameter $\alpha = M_S H/2K_U$. The equilibrium angle of the magnetization is then determined by $\partial \alpha/\partial \theta=0$, i.e., $\sin 2\theta + 2\alpha \sin(\theta - \phi)=0$. The equation provides multi-case solutions of $\theta(\alpha, \phi)$ depending on the initial condition, as given by Ref. 13 or readily solved by use of MATHEMATICA. For heuristic purpose, we solely focus here on the case of $\phi=0$ and $\theta=0$, from which the other case of $\phi \sim \pi$ and $\theta \sim \pi$ can be easily derived by the symmetry argument. For small $\phi$ and $\theta$, the solution can be written by the Fourier series expansion as $\cos \theta = 1 + A_2 \phi^2 + A_4 \phi^4 + A_6 \phi^6 + \ldots$, where the Fourier coefficients are given by

$$
A_2(\alpha) = \frac{\alpha^2}{2 \times (1 + \alpha)},
$$

$$
A_4(\alpha) = \frac{\alpha^2(4 - 15 \alpha^2 + \alpha^4)}{4 \times (1 + \alpha)^2},
$$

$$
A_6(\alpha) = \frac{\alpha^2(16 - 54 \alpha - 300 \alpha^2 + 260 \alpha^3 + 945 \alpha^4 - 174 \alpha^5 + \alpha^6)}{6 \times (1 + \alpha)^3}.
$$

Figure 1 shows the Fourier coefficients with respect to $\alpha$. As clearly seen from the figure, the higher order coefficients are negligibly smaller than $A_2$. It is thus quite reasonable to put the asymptotic solution as

$$
\cos \theta \approx 1 + \frac{(M_S H)^2}{2(2K_U + M_S H)^2} \phi^2,
$$

for fairly wide range of $\phi$. In the experiments we restrict $|\phi| \leq \pi/6$ to avoid the switching of the magnetization. We confirm that inclusion/exclusion of the fourth order term $A_4$ does not make any noticeable change in comparison with the experimental accuracy.

The magnetization angle $\theta$ is experimentally measured by means of the Hall effect. In the measurement setup, four wires are bonded at each corner of square-cut samples. One
pair of diagonal corners is connected to the current source, and the other pair is used for the voltage probe suggested as van der Pauw method. However, in our experiment, it is not important to measure the exact value of the Hall voltage, so the electric connections make no significant problems. Figure 2 shows the wire connections of the samples. When a constant current of 10 mA flows, transverse electric voltage is induced by the Hall effect. The Hall voltage \( V_H \) is expressed as 
\[
V_H = R_s I_H + R_s I_M, \quad H = H \cos \phi, \quad M_s = M_s \cos \theta, \quad R_s \text{ is the ordinary Hall coefficient, } R_s \text{ is the extraordinary Hall coefficient, and } I \text{ is the current flow in the sample.}
\]
The voltage probes thus detect the out-of-plane components of the magnetization and the external field, which are proportional to \( \cos \theta \) and \( \cos \phi \), respectively.

To measure \( \phi \)-dependence of the magnetization angle \( \theta \), a constant external magnetic field is applied normal to the film, and then, the film is rotated by \( \phi \) with the rotation axis, parallel to the current flow. In this method, the ordinary Hall voltage can be easily subtracted. During one rotation of \( \phi \) from \(-180^\circ\) to \(180^\circ\), the ordinary Hall effect generates the signal proportional to \( H \cos \phi \). The amplitude of the ordinary Hall voltage linearly depends on the external field. Hence, we can exactly subtract the ordinary Hall voltage by measuring with different applied fields. However, in realistic cases, the ordinary Hall effect is negligible because it is much smaller than the extraordinary Hall effect due to \( R_s \ll R_s \) and \( H < M_s \). Finally, one readily calibrates \( \cos \theta \) after normalization with the relation 
\[
\cos \theta = \frac{V_H - (V_{H_{\max}} + V_{H_{\min}})/2}{(V_{H_{\max}} - V_{H_{\min}})/2}.
\]
\( V_{H_{\max}} \) and \( V_{H_{\min}} \) are the maximum and minimum values of \( V_H \) in the one round rotation measurement.

The present measurement technique is applied to CoFe/Pt multilayer films. 50 Å Ta/25 Å Pt/(10 Å Co90Fe10)/10 Å Pt\(_n\) films are deposited on Si substrate with natural SiO\(_2\) layer using d.c.-magnetron sputtering with changing the number of repeats \( n \) from 1 to 5. From the magnetooptical Kerr effect measurement, all the films are revealed to exhibit squared out-of-plane hysteresis loops, evidencing the strong perpendicular magnetic anisotropy. The coercive field is monotonically increased with increasing \( n \) as listed in Table I. The saturation magnetization \( M_S \) per Co volume is 2.0 ± 0.2 T for all the films, measured by the alternating gradient magnetometer.

Figure 3 plots the extraordinary Hall voltage \( V_H \) with respect to the sweeping angle \( \phi \) for the film with \( n=2 \). \( V_H \) is jump up and down at the angles \(-90^\circ\) and \(90^\circ\) due to the magnetization reversal shown in the inset. The normalized value of \( \cos \theta \) is shown in the right side of the plots. The rotation sense does not induce any noticeable changes near the angles 0° and 180°, which confirms that the magnetization process in this angle range is fairly reversible and therefore compatible with the Stoner–Wohlfarth model. \( \cos \theta \) is well fitted to the parabolic function given by Eq. (2). From the best fit shown by the black line in Fig. 3, one can obtain the Fourier coefficient \( A_2 \). The coefficient is then converted to the anisotropy field \( H_K \) by

\[
H_K = \frac{2K_U}{M_S} = H \frac{1 - \sqrt{2A_2}}{\sqrt{2A_2}}.
\]

or to the uniaxial anisotropy \( K_U \) if the value of \( M_S \) is provided.

The Fourier coefficient \( A_2 \) is measured with respect to the external field \( H \). Figure 4 shows \( A_2(H) \) for the films with different \( n \)—(a) 2, (b) 3, (c) 4, and (d) 5, respectively. The

\[
FIG. 1. The Fourier coefficients \( A_n \) with respect to \( \alpha \).
\]

\[
FIG. 2. The measurement geometry of the sample.
\]

\[
FIG. 3. The extraordinary Hall voltage \( V_H \) and normalized \( \cos \theta \) with respect to the sweeping angle \( \phi \) for the film with \( n=2 \). The black line is the best fit with Eq. (2) by \( \chi^2 \)-square fitting. The magnetic field strength \( H \) is 200 mT.
\]

\[
TABLE I. The anisotropy field \( H_K \), perpendicular magnetic anisotropy \( K_U \), and the coercive field \( H_C \) of the CoFe/Pt multilayers with different numbers of repeat \( n \).
\]

<table>
<thead>
<tr>
<th>( n )</th>
<th>( H_K ) (T)</th>
<th>( K_U ) ((10^6 , J/m^3))</th>
<th>( H_C ) (mT)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.39 ± 0.16</td>
<td>1.11 ± 0.25</td>
<td>11.3</td>
</tr>
<tr>
<td>2</td>
<td>1.37 ± 0.03</td>
<td>1.09 ± 0.13</td>
<td>27.2</td>
</tr>
<tr>
<td>3</td>
<td>1.28 ± 0.05</td>
<td>1.02 ± 0.15</td>
<td>29.5</td>
</tr>
<tr>
<td>4</td>
<td>1.20 ± 0.02</td>
<td>0.96 ± 0.12</td>
<td>36.0</td>
</tr>
<tr>
<td>5</td>
<td>1.13 ± 0.01</td>
<td>0.90 ± 0.10</td>
<td>36.3</td>
</tr>
</tbody>
</table>
solid line is the best fit by 

\[ A_2(H) = \frac{H^2}{2(H + H_{K})^2} \]

given by Eq. (1). The absolute conformity verifies the validity of the present measurement technique, as well as provides fairly reliable values of \( H_K \). The experimental values of \( H_K \) and \( K_U \) are summarized in Table I. The perpendicular magnetic anisotropy is found to be monotonically decreased with increasing \( n \). It is possibly ascribed to the accumulation of the CoFe/Pt interfacial irregularities such as atomic misfits, defects, dislocations, and crystalline misorientations with increasing the number of layers.

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