ORIGINAL ARTICLE

Accurate analysis of harmonic Hall voltage measurement for spin–orbit torques

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An accurate method is developed to extract the spin–orbit (SO) effective fields by analyzing harmonic Hall voltage measurements and deriving detailed analytical equations that consider both the z component of the applied magnetic field and the second-order perpendicular magnetic anisotropy. The method is tested by analyzing the results of a macrospin simulation. The SO effective fields that are extracted from the analysis are consistent with the input SO effective fields that are used in the macrospin simulation over the entire range of the polar magnetization angle and for a wide range (0–2) of the ratio of the planar to the anomalous Hall voltage considered in this study. The accuracy of the proposed method is demonstrated via a systematic study that involves comparing its results with those of a conventional analytical method.

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INTRODUCTION

In-plane currents in a nonmagnetic (NM)/ferromagnetic (FM) bilayer nanostructure can generate a torque due to the spin–orbit (SO) coupling, which is known as the SO torque (SOT). SOT is sufficient for reversing the magnetization in the FM layer.¹ Numerous studies have been conducted to identify the principal mechanism of the SOT as either the spin Hall effect (SHE) in the NM layer^{2–4} or the interfacial SO coupling—frequently referred to as the Rashba effect—at the NM/FM interface.^{5–12} In a system in which the NM/FM interface is perpendicular to the z axis and the in-plane currents flow along the x axis, spin currents that are polarized along the y axis are generated in the system based on the SHE induced by a bulk SO coupling in the NM layer. The spin currents are injected into the adjacent FM layer, which causes torque to be transferred to the magnetization of the FM layer. The SHE-induced SOT generates a strong damping-like torque ($T_{\text{DL}} \propto \mathbf{m} \times \mathbf{m} \times \mathbf{y}$) and a weak field-like torque ($T_{\text{FL}} \propto \mathbf{m} \times \mathbf{y}$).^{13,14} Theoretically, the strength of the SHE-induced SOT is known to be independent of the magnetization direction of the FM layer. For an interfacial SO coupling-induced SOT, spins that are polarized along the y axis accumulate due to the broken inversion symmetry at the NM/FM interface. Direct exchange coupling between the magnetization of the FM layer and the accumulated spins generates a strong T_{FL} but a weak T_{DL} .^{7,15–18} Unlike the strength of the SHE-induced SOT, the interfacial SO coupling-induced SOT is known to depend on the magnetization direction of the FM layer.^{19–21} In these two cases, both the SHE and the interfacial SO coupling qualitatively induce the same torque on the FM layer. To identify the dominant mechanism of the SOT, a quantitative analysis of the values of T_{DL} and T_{FL} over an extensive range of magnetization angles is required.^{19–21}

The harmonic Hall voltage measurement method is a useful approach for quantifying the effective fields of T_{DL} and T_{FL} that originate from the SOT.²² This method is particularly suitable for identifying the angular dependence of the SOT that acts on the FM layer with perpendicular magnetization.^{19,20} Several corrections are required for an accurate analysis of the measured results, including the measured results for the planar Hall effect (PHE),^{20,23} the out-of-plane component of the external magnetic field²⁰ and the anomalous Nernst effect.^{20,24} In the harmonic Hall voltage measurement, the second harmonic voltage ($V^{2\omega}$) consists of two major components: anomalous Hall voltage and planar Hall voltage (denoted as V_{AHE} and V_{PHE} , respectively).^{20,23} When an external magnetic field ($\mathbf{H}_{\text{ext}}$) is applied along the longitudinal (x) direction, the $V^{2\omega}$ values from the AHE and PHE are proportional to T_{DL} and T_{FL} , respectively. Under a transverse (y) $\mathbf{H}_{\text{ext}}$, however, these values are proportional to T_{FL} and T_{DL} , respectively. This necessitates the use of an analytical solution that is based on Cramer's rule to separate T_{FL} and T_{DL} .²³ This analytical solution has been successfully applied to only a system in which $V_{\text{PHE}} < V_{\text{AHE}}$. For a system in which $V_{\text{PHE}} > V_{\text{AHE}}$, such as the W/CoFeB/MgO trilayer structure,²⁵ a divergence occurs in the solution, which makes it extremely difficult to analyze the measurement results.

This problem can be overcome by incorporating some necessary corrections into the analysis of the measurement results, including measurement results for the out-of-plane component of $\mathbf{H}_{\text{ext}}$. As coherent magnetization rotation is an important requirement in the analysis of harmonic Hall voltage measurement results, $\mathbf{H}_{\text{ext}}$ is usually applied along the direction tilted slightly (4° – 15°) from the basal plane (x – y plane).²⁰ In this condition, the z component of $\mathbf{H}_{\text{ext}}$ has a nonzero value, although it has been neglected to simplify the analysis. This assumption is reasonable in the low- $\mathbf{H}_{\text{ext}}$ range, where the

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magnetization direction is close to the z axis and the perpendicular magnetic anisotropy (PMA) field is consequently dominant over the z component of $\mathbf{H}_{\text{ext}}$.²³ The simplifying assumption, however, is not valid in the high- $\mathbf{H}_{\text{ext}}$ range, where the magnetization direction deviates considerably from the z axis, with a resultant reduction in the PMA field. As a result, the PMA field loses its dominance over the z component of $\mathbf{H}_{\text{ext}}$. Several attempts have been made to include the z component of $\mathbf{H}_{\text{ext}}$, which is obtained by repetitively solving equilibrium torque equations until the desired convergence is achieved (the recursive method).^{19,20} However, this method is complicated, and it has not been validated for systems with $V_{\text{PHE}} > V_{\text{AHE}}$. Unwanted voltages, which originate from thermoelectric effects such as the anomalous Nernst effect, should be eliminated from the harmonic signals. Although several methods have been proposed for this purpose,^{20,24} erasing all artificial signals remains difficult. Another important issue that has not been considered is the inclusion of the second-order PMA in an analysis of the harmonic Hall voltage measurement results. The inclusion of the second-order PMA is considered important because many PMA materials exhibit second-order PMA, whose strength is comparable to that of the first-order PMA in many cases.^{26,27}

In this study, two corrections—one for the z component of $\mathbf{H}_{\text{ext}}$ and the other for the second-order PMA—are considered in the analysis of the harmonic Hall voltage measurement results. All related analytical equations are described. Both conventional and refined analytical methods are used to analyze the results of the macrospin simulation, which mimics the harmonic Hall voltage measurement by numerically solving the Landau–Lifshitz–Gilbert equation.^{28,29} The accuracy of these two analytical methods is tested by comparing the input SO effective fields for the macrospin simulation with the input SO effective fields calculated by analytical methods. To test the refined analytical method, systems are examined over a wide ratio R , which is defined as $V_{\text{PHE}}/V_{\text{AHE}}$. A similar comparative study that analyzes the experimental results of harmonic Hall voltage measurements for a Pt/Co/MgO structure was also performed.

MATERIALS AND METHODS

Analytical solutions for conventional approach

When an in-plane AC current with frequency ω ($I_{\text{AC}} = I_0 \sin \omega t$) is applied to an NM/FM bilayer structure, the angle between the z axis and the magnetization of the FM layer (θ_M) and the angle between the x axis and the orthographic projection of the magnetization on the x - y plane (φ_M) oscillate as $\theta_M(t) = \theta_M^0 + \Delta\theta_M \sin \omega t$ and $\varphi_M(t) = \varphi_M^0 + \Delta\varphi_M \sin \omega t$. Here the superscript 0 and the symbol Δ denote the value in the absence of I_{AC} and the amplitude of the related angles, respectively. The total energy equation for the NM/FM bilayer structure can be expressed as follows:

$$E_{\text{tot}} = -K_1^{\text{eff}} \cos^2 \theta_M - K_2 \cos^4 \theta_M - M_S \mathbf{m} \cdot (\mathbf{H}_{\text{ext}} + \Delta \mathbf{H}), \quad (1)$$

$$\mathbf{m} \equiv (\sin \theta_M \cos \varphi_M, \sin \theta_M \sin \varphi_M, \cos \theta_M), \quad (2)$$

$$\mathbf{H}_{\text{ext}} \equiv H_{\text{ext}} (\sin \theta_H \cos \varphi_H, \sin \theta_H \sin \varphi_H, \cos \theta_H), \quad (3)$$

$$\Delta \mathbf{H} \equiv \Delta H_{\text{DL}} \mathbf{m} \times \hat{y} - \Delta H_{\text{FL}} \hat{y}. \quad (4)$$

Here K_1^{eff} is the effective first-order PMA energy density that considers the demagnetizing term, that is, $K_1^{\text{eff}} = K_1 - N_d M_S^2/2$ (K_1 , N_d and M_S are the first-order PMA energy density, demagnetizing factor and saturation magnetization, respectively).³⁰ K_2 is the second-order PMA energy density.³⁰ $\mathbf{m}$ is the unit vector of magnetization. The effective magnetic field ($\Delta \mathbf{H}$) induced by the maximum value of the in-plane AC current (I_0) is composed of the damping-like effective field (ΔH_{DL}) and the field-like effective field (ΔH_{FL}). θ_H and φ_H are the polar angle and azimuthal angle of $\mathbf{H}_{\text{ext}}$, respectively. Given that the in-plane anisotropy is negligibly small over the PMA field, φ_M^0 is assumed to be

identical to φ_H . The values of $\Delta\theta_M$ and $\Delta\varphi_M$ can be analytically expressed as follows (refer to Supplementary Equations (S1)–(S13) for a detailed derivation):

$$\Delta\theta_M = \frac{\Delta H_{\text{DL}} \cos \varphi_H + \Delta H_{\text{FL}} \cos \theta_M^0 \sin \varphi_H}{H_K^{\text{eff}} \cos 2\theta_M^0 - H_{K,2} \sin \theta_M^0 \sin 3\theta_M^0 + H_{\text{ext}} \cos (\theta_M^0 - \theta_H)}, \quad (5)$$

$$\Delta\varphi_M = \frac{\Delta H_{\text{DL}} \cos \theta_M^0 \sin \varphi_H - \Delta H_{\text{FL}} \cos \varphi_H}{H_{\text{ext}} \sin \theta_H}. \quad (6)$$

Here H_K^{eff} and $H_{K,1}^{\text{eff}}$ are the effective PMA field and the effective first-order PMA field, respectively, and $H_{K,2}$ is the second-order PMA field. These parameters are defined as follows: $H_K^{\text{eff}} \equiv H_{K,1}^{\text{eff}} + H_{K,2}$; $H_{K,1}^{\text{eff}} \equiv 2K_1^{\text{eff}}/M_S$; $H_{K,2} \equiv 4K_2/M_S$. Note that Equations (5) and (6) are identical to the analytical expressions derived by Hayashi *et al.*²³ when $H_{K,2} = 0$. If the values of $\Delta\theta_M$ and $\Delta\varphi_M$ are sufficiently small, the components of the $\mathbf{m}$ vector can be approximated in the form $m(t) = m^0 + (2\Delta m) \sin \omega t$:

$$m_x \approx \sin \theta_M^0 \cos \varphi_H + (\Delta\theta_M \cos \theta_M^0 \cos \varphi_H - \Delta\varphi_M \sin \theta_M^0 \sin \varphi_H) \sin \omega t, \quad (7)$$

$$m_y \approx \sin \theta_M^0 \sin \varphi_H + (\Delta\theta_M \cos \theta_M^0 \sin \varphi_H + \Delta\varphi_M \sin \theta_M^0 \cos \varphi_H) \sin \omega t, \quad (8)$$

$$m_z \approx \cos \theta_M^0 - \Delta\theta_M \sin \theta_M^0 \sin \omega t. \quad (9)$$

Both the anomalous Hall voltage and planar Hall voltage contribute to the measured Hall voltage: $V_H = I_{\text{AC}} R_H = I_{\text{AC}} R_{\text{AHE}} m_z + I_{\text{AC}} R_{\text{PHE}} m_x m_y$.^{20,31,32} Here R_{AHE} and R_{PHE} are the anomalous and planar Hall resistance, respectively. With the application of I_{AC} , the m values oscillate as given in Equations (7)–(9) with the resultant expressions for Hall voltages as follows:

$$V_H = (V_{\text{AHE}} m_z + V_{\text{PHE}} m_x m_y) \sin \omega t = V^{1\omega} \sin \omega t - V^{2\omega} \cos 2\omega t, \quad (10)$$

$$V_x^{1\omega} = V_y^{1\omega} = V_{\text{AHE}} \cos \theta_M^0, \quad (11)$$

$$V_x^{2\omega} = -\frac{V_{\text{AHE}}}{2} [\sin \theta_M^0 \Delta\theta_M - R \sin^2 \theta_M^0 \Delta\varphi_M], \quad (12)$$

$$V_y^{2\omega} = -\frac{V_{\text{AHE}}}{2} [\sin \theta_M^0 \Delta\theta_M + R \sin^2 \theta_M^0 \Delta\varphi_M]. \quad (13)$$

Here the following relations exist: $V_{\text{AHE}} = I_0 R_{\text{AHE}}$ and $V_{\text{PHE}} = I_0 R_{\text{PHE}}$. The subscripts x and y indicate that the harmonic Hall voltages are measured at $\varphi_H = 0^\circ$ and $\varphi_H = 90^\circ$, respectively. The first harmonic Hall voltage ($V^{1\omega}$) contains information about the θ_M^0 value, and the second harmonic Hall voltage ($V^{2\omega}$) contains information about the $\Delta\theta_M$ and $\Delta\varphi_M$ values. The conventional analytical solution considers only the case in which the magnetization direction has slightly deviated from the z axis ($\theta_M^0 \approx 0^\circ$). In this case, the z component of $\mathbf{H}_{\text{ext}}$ is negligibly small over the PMA field along the same direction ($H_{\text{ext}} \cos \theta_H \ll H_K^{\text{eff}} \cos \theta_M^0$), and therefore, the assumption of $\sin \theta_M^0 = H_{\text{ext}}/H_K^{\text{eff}}$ in the conventional solution is reasonable.^{8,23,33} Note that the contribution due to $H_{K,2}$ is also negligible at $\theta_M^0 \approx 0^\circ$ (Equation (5)). With this assumption, $V^{1\omega}$ and $V^{2\omega}$ can be rewritten as follows:

$$V_x^{1\omega} = V_y^{1\omega} = V_{\text{AHE}} \sqrt{1 - \left(\frac{H_{\text{ext}}}{H_K^{\text{eff}}} \right)^2}, \quad (14)$$

$$V_x^{2\omega} = \frac{V_{\text{AHE}} H_{\text{ext}}}{2(H_K^{\text{eff}})^2} \left[\frac{\Delta H_{\text{DL}}}{1 - (H_{\text{ext}}/H_K^{\text{eff}})^2} - R \Delta H_{\text{FL}} \right], \quad (15)$$

$$V_y^{2\omega} = \frac{V_{\text{AHE}} H_{\text{ext}}}{2(H_K^{\text{eff}})^2} \sqrt{1 - \left(\frac{H_{\text{ext}}}{H_K^{\text{eff}}} \right)^2} \left[\frac{\Delta H_{\text{FL}}}{1 - (H_{\text{ext}}/H_K^{\text{eff}})^2} - R \Delta H_{\text{DL}} \right]. \quad (16)$$

The second harmonic Hall voltages, as given in Equations (15) and (16), are composed of two terms that contain ΔH_{DL} and ΔH_{FL} . When the R ratio is negligibly small, the values of ΔH_{DL} and ΔH_{FL} can be obtained using the

following T ratios:

$$T_x \equiv -\frac{2V_x^{2\omega}}{(\partial V_x^{1\omega}/\partial H_{\text{ext}})} \left(\frac{V_x}{V_{\text{AHE}}} \right) = A_0 \Delta H_{\text{DL}} - B_0 \Delta H_{\text{FL}}, \quad (17)$$

$$T_y \equiv -\frac{2V_y^{2\omega}}{(\partial V_y^{1\omega}/\partial H_{\text{ext}})} = B_0 \Delta H_{\text{DL}} - A_0 \Delta H_{\text{FL}}, \quad (18)$$

$$A_0 \equiv 1, B_0 \equiv R \left(1 - \left(\frac{H_{\text{ext}}}{H_{\text{K}}^{\text{eff}}} \right)^2 \right). \quad (19)$$

At $R=0$, the values of T_x and T_y are identical to the values of ΔH_{DL} and $-\Delta H_{\text{FL}}$, respectively.⁸ When the R ratio becomes comparatively large, T_x and T_y should be corrected using Cramer's rule.²³

$$\begin{pmatrix} \Delta H_{\text{DL}} \\ \Delta H_{\text{FL}} \end{pmatrix} = \frac{1}{B_0^2 - A_0^2} \begin{pmatrix} -A_0 & B_0 \\ -B_0 & A_0 \end{pmatrix} \begin{pmatrix} T_x \\ T_y \end{pmatrix}. \quad (20)$$

In Equation (20), ΔH_{DL} and ΔH_{FL} can be calculated if the determinant $B_0^2 - A_0^2$ is not 0. If $B_0^2 - A_0^2$ is 0, it is not possible to obtain the individual values of ΔH_{DL} and ΔH_{FL} ; it is only possible to obtain the relation $T_x = T_y = \Delta H_{\text{DL}} - \Delta H_{\text{FL}}$ (Equations (17)–(19)).

Analytical solutions for refined approach

The assumption of $H_{\text{ext}} \cos \theta_H \ll H_{\text{K}}^{\text{eff}} \cos \theta_M^\circ$ is not valid at high H_{ext} values. In this case, the θ_H value is not negligible, and $\Delta \theta_M$ and $\Delta \varphi_M$ (Equations (5) and (6)) must be substituted into Equations (12) and (13) to obtain the expression for $V^{2\omega}$:

$$V_x^{2\omega} = \frac{V_{\text{AHE}}}{2} [A_1 \Delta H_{\text{DL}} - B_1 \Delta H_{\text{FL}}], \quad (21)$$

$$V_y^{2\omega} = -\frac{V_{\text{AHE}} \cos \theta_M^\circ}{2} [B_1 \Delta H_{\text{DL}} - A_1 \Delta H_{\text{FL}}], \quad (22)$$

$$A_1 \equiv \frac{\sin \theta_M^\circ}{H_{\text{K}}^{\text{eff}} \cos 2\theta_M^\circ - H_{\text{K},2} \sin \theta_M^\circ \sin 3\theta_M^\circ + H_{\text{ext}} \cos (\theta_M^\circ - \theta_H)}, \quad (23)$$

$$B_1 \equiv \frac{R \sin^2 \theta_M^\circ}{H_{\text{ext}} \sin \theta_H}. \quad (24)$$

Considering that $V_x^{1\omega} = V_y^{1\omega} = V_{\text{AHE}} \cos \theta_M^\circ$ (Equation (11)), the G ratios, which correspond to the T ratios in the conventional approach, can be defined as follows:

$$G_x \equiv \frac{2V_x^{2\omega}}{V_{\text{AHE}}} = A_1 \Delta H_{\text{DL}} - B_1 \Delta H_{\text{FL}}, \quad (25)$$

$$G_y \equiv -\frac{2V_y^{2\omega}}{V_y^{1\omega}} = B_1 \Delta H_{\text{DL}} - A_1 \Delta H_{\text{FL}}. \quad (26)$$

Similar to the conventional analytical equations, the refined equations also need to be solved using Cramer's rule (as given in Equation (20)).

$$\begin{pmatrix} \Delta H_{\text{DL}} \\ \Delta H_{\text{FL}} \end{pmatrix} = \frac{1}{B_1^2 - A_1^2} \begin{pmatrix} -A_1 & B_1 \\ -B_1 & A_1 \end{pmatrix} \begin{pmatrix} G_x \\ G_y \end{pmatrix}. \quad (27)$$

RESULTS

Conventional analysis

The conventional analytical method is used to analyze the results of the macrospin simulation. Figure 1a and b shows the results for $V^{1\omega}$ as a function of H_{ext} in two different systems: (a) $H_{\text{K},1}^{\text{eff}} = 5$ kOe and $H_{\text{K},2} = 0$ kOe; and (b) $H_{\text{K},1}^{\text{eff}} = 5$ kOe and $H_{\text{K},2} = -1$ kOe. Two sets of results are shown in Figure 1a and b: the first set is obtained from the macrospin simulation (squares), and the second set is obtained from Equation (14), which is based on the conventional assumption of $\sin \theta_M^\circ = H_{\text{ext}}/H_{\text{K}}^{\text{eff}}$ (dashed lines). For the macrospin simulation, the

following parameters are applied: $\Delta H_{\text{DL}} = -50$ Oe; $\Delta H_{\text{FL}} = -100$ Oe; $\theta_H = 86^\circ$; and $V_{\text{AHE}} = 1$ mV. Refer to Supplementary Figure S1 for a detailed description of the macrospin simulation. Agreement between the results obtained from the macrospin simulation and the results obtained from Equation (14) based on the conventional analytical method, which is the focus of this study, is only obtained in the low- H_{ext} range. In the high- H_{ext} range, the deviation is very large, which indicates the limited validity of conventional solutions.

Figure 1c and d shows the analytical results for $B_0^2 - A_0^2$ calculated from Equation (19) as a function of H_{ext} at two different R values of 0.3 (red lines) and 1.75 (blue lines) ($R = V_{\text{PHE}}/V_{\text{AHE}}$). The results in Figure 1c pertain to the system with $H_{\text{K},1}^{\text{eff}} = 5$ kOe and $H_{\text{K},2} = 0$, and the results in Figure 1d pertain to the system with $H_{\text{K},1}^{\text{eff}} = 5$ kOe and $H_{\text{K},2} = -1$ kOe. The $H_{\text{K}}^{\text{eff}}$ values for both systems are also indicated in the figures. The detailed equation for $B_0^2 - A_0^2$ is rewritten for clarity:

$$B_0^2 - A_0^2 = R^2 \left(1 - \left(\frac{H_{\text{ext}}}{H_{\text{K}}^{\text{eff}}} \right)^2 \right)^2 - 1. \quad (28)$$

Recalling that $H_{\text{ext}}/H_{\text{K}}^{\text{eff}}$ is approximated with $\sin \theta_M^\circ$, we note that the results for $B_0^2 - A_0^2$ at $H_{\text{ext}} > H_{\text{K}}^{\text{eff}}$ have no physical meaning. According to Equation (28), the $B_0^2 - A_0^2$ value decreases from $R^2 - 1$ to -1 as the H_{ext} value increases from 0 to $H_{\text{K}}^{\text{eff}}$. The determinant $B_0^2 - A_0^2$ always has a negative value at $R < 1$. At $R \geq 1$, however, the determinant can have both positive values and negative values over the H_{ext} range of $0 - H_{\text{K}}^{\text{eff}}$, which indicates the occurrence of a crossover ($B_0^2 - A_0^2 = 0$) at a certain H_{ext} value. This feature is visible in the results shown in Figure 1c and d. In both systems, that is, with $H_{\text{K},2} = 0$ and -1 kOe, the $B_0^2 - A_0^2$ value is always negative at $R = 0.3$; at $R = 1.75$, it initially has a positive value, after which it passes through 0 and then has a negative value. The crossovers occur at 3.3 and 2.6 kOe for the systems with $H_{\text{K},2} = 0$ kOe and $H_{\text{K},2} = -1$ kOe, respectively. Recalling that $T_x = T_y = \Delta H_{\text{DL}} - \Delta H_{\text{FL}}$ when the determinant is 0, an H_{ext} value should exist at $T_x = T_y$ when $R > 1$.

Figure 2a–f shows the results for $V_x^{2\omega}$ and $V_y^{2\omega}$ (a and b) and T_x and T_y (c and d) as functions of H_{ext} and the results for ΔH_{DL} and ΔH_{FL} (e and f) as functions of θ_M° for two different systems, that is, with $H_{\text{K},2} = 0$ (solid lines) and $H_{\text{K},2} = -1$ kOe (dashed lines). The left (Figure 2a, c and e) and right panels (Figure 2b, d and f) show the results for $R = 0.3$ and 1.75, respectively. The results for $V^{2\omega}$ were obtained by the macrospin simulation, and the results for T_x and T_y (Equations (17) and (18)) and ΔH_{DL} and ΔH_{FL} (Equation (20)) were analytically calculated using the simulation results. As shown in Figure 2a and b, the sign of $V_x^{2\omega}$ for a small R value of 0.3 is negative, whereas the sign of $V_x^{2\omega}$ for a large R value of 1.75 is positive. Of the two major contributions of V_{AHE} and V_{PHE} to $V^{2\omega}$, the sign of the former is negative, but that of the latter is positive. Equations (15) and (16) predict this behavior (the opposite signs of V_{AHE} and V_{PHE}) and explain that the $V_y^{2\omega}$ value for $R = 1.75$ is higher than that for $R = 0.3$.

The results for $V_x^{2\omega}$ and $V_y^{2\omega}$ and their variation with R have a critical effect on T_x and T_y . At $R = 0.3$, the signs of T_x and T_y are opposite because the signs of $V_x^{2\omega}$ and $V_y^{2\omega}$ are the same, indicating that there are no H_{ext} values at which $T_x = T_y$ in both systems, that is, for $H_{\text{K},2} = 0$ kOe and $H_{\text{K},2} = -1$ kOe (Figure 2c). These results are consistent with the results for $B_0^2 - A_0^2$ (Figure 1c and d). The values of T_x and T_y are the same at a specific H_{ext} value, at which $B_0^2 - A_0^2 = 0$ (Equation (19)). For $R = 1.75$, the signs of T_x and T_y are the same because the signs of $V_x^{2\omega}$ and $V_y^{2\omega}$ are opposite (Figure 2d). There are H_{ext} values for which $T_x = T_y$ exist in both systems, that is, with $H_{\text{K},2} = 0$ kOe and $H_{\text{K},2} = -1$ kOe. The positions, however, differ from those for which $B_0^2 - A_0^2 = 0$. The H_{ext} values in

Figure 1 Results for $V^{1\omega}$ (a, b) and $B_0^2 - A_0^2$ (c, d) as functions of H_{ext} for systems with $H_{K,1}^{\text{eff}} = 5$ kOe and $H_{K,2} = 0$ kOe (left panels; a, c) and $H_{K,1}^{\text{eff}} = 5$ kOe and $H_{K,2} = -1$ kOe (right panels; b, d). For the macrospin simulation, the following parameters are used: $\Delta H_{\text{DL}} = -50$ Oe; $\Delta H_{\text{FL}} = -100$ Oe; $\theta_H = 86^\circ$; and $V_{\text{AHE}} = 1$ mV. In a, b, two sets of results are shown: one set of results from the macrospin simulation (squares) and one set from Equation (14), which is based on the conventional analytical method (dashed lines). In c, d, two sets of results, both of which are obtained by the conventional analytical method, are shown for $R = 0.3$ (red lines) and $R = 1.75$ (blue lines).

the former case are 3.6 and 3.8 kOe for the systems with $H_{K,2} = 0$ and $H_{K,2} = -1$ kOe, respectively, whereas those in the latter case are 3.3 and 2.6 kOe, respectively. These deviations occur because the determinant poorly reflects the behavior of the first harmonic.

The inappropriate determinant, that is, $B_0^2 - A_0^2$, causes large errors in the SO effective fields, as shown in Figure 2e and f. Recalling that the input SO effective fields are $\Delta H_{\text{DL}} = -50$ Oe and $\Delta H_{\text{FL}} = -100$ Oe, we consider the results at $R = 0.3$ (Figure 2e) to be reliable in the θ_M° range from 0° to the angles that correspond to $H_{\text{ext}} = H_K^{\text{eff}}$. These angles are 61° and 52° when $H_{K,2} = 0$ kOe and $H_{K,2} = -1$ kOe, respectively. Beyond these two angles, which are indicated by the vertical and horizontal lines in Figure 2e and f, respectively, the output SO effective fields start to deviate from the input values. The indicated regions end not at 90° but at $\sim 82^\circ$ as the $\mathbf{m}$ vector is not completely aligned along the x or y axis, even for $H_{\text{ext}} = 10$ kOe (Figure 1a and b). The output SO effective fields reveal a divergence, which is physically meaningless at $\theta_M^\circ = \sim 81^\circ$ ($H_{K,2} = 0$ kOe) and $\sim 75^\circ$ ($H_{K,2} = -1$ kOe). The deviations are very large at $R = 1.75$ (Figure 2f). For the system with $H_{K,2} = 0$ kOe, divergence occurs even at $\sim 37^\circ$, which is not located in the region of physical insignificance (indicated by vertical lines). A similar behavior is observed for the system with $H_{K,2} = -1$ kOe, where the divergence occurs at $\sim 32^\circ$. These divergences are attributed to the misallocation of the H_{ext} value, at which $B_0^2 - A_0^2 = 0$ (Figure 1c and d). The occurrence of the additional divergences significantly limits the reliability of the conventional analytical method for both systems, that is, with $H_{K,2} = 0$ kOe and $H_{K,2} = -1$ kOe, as shown in Figure 2f.

Refined analysis

The main reason behind the unreliable results obtained from the conventional analysis is the determinant, which poorly describes the behavior of the first harmonic Hall voltage. For an accurate evaluation of the determinant, the refined analysis begins by determining the relation between θ_M° and H_{ext} , which can be obtained using the equation $\theta_M^\circ = \cos^{-1}(V^{1\omega}/V_{\text{AHE}})$ or the total energy equation³⁴

Figure 2 Results for $V_x^{2\omega}$ and $V_y^{2\omega}$ (a, b) and T_x and T_y (c, d) as functions of H_{ext} and those for ΔH_{DL} and ΔH_{FL} (e, f) as functions of θ_M° . The results for $V^{2\omega}$ are obtained by a macrospin simulation, and those for T_x , T_y , ΔH_{DL} and ΔH_{FL} are obtained by analysis of the simulation results using a conventional analytical method. Results are obtained for the systems with $H_{K,2} = 0$ kOe (solid lines) and $H_{K,2} = -1$ kOe (dashed lines). The left panels (a, c, e) show the results for $R = 0.3$, whereas the right panels (b, d, f) show the results for $R = 1.75$. In e, f, the regions filled with vertical lines and horizontal lines indicate the θ_M° range of no physical significance for the systems with $H_{K,2} = 0$ and $H_{K,2} = -1$ kOe, respectively.

(Equation (1)). The results for $V^{1\omega}$ as a function of H_{ext} (Figure 1a and b) can be applied to obtain the relation. Figure 3a and b shows the results for the determinant $B_1^2 - A_1^2$ as a function of the H_{ext} obtained from the refined analysis (Equations (23) and (24)) for the systems with $H_{K,2} = 0$ kOe and $H_{K,2} = -1$ kOe, respectively. The solid lines in Figure 3a and b indicate the results for $B_1^2 - A_1^2$, as calculated using $H_{K,2} = 0$ and $H_{K,2} = -1$ kOe from the macrospin simulation and the refined analysis, respectively. Using the relation between θ_M° and H_{ext} in the refined analysis enables the behavior of $V^{1\omega}$ to be duly reflected in the determinant. At $R = 1.75$, the H_{ext} values for which $B_1^2 - A_1^2 = 0$ are 3.6 and 3.8 kOe for the systems with $H_{K,2} = 0$ and $H_{K,2} = -1$ kOe, respectively; these H_{ext} values are identical to those obtained at $T_x = T_y$ (Figure 2d). To apply the behavior of $V^{1\omega}$ to the determinant, the determinant $B_1^2 - A_1^2$ needs to be calculated with a precise value of $H_{K,2}$, which was used in the macrospin simulation. To demonstrate the importance of the inclusion of $H_{K,2}$, the determinants were also calculated by disregarding $H_{K,2}$ (even though the actual $H_{K,2}$ value of the system is -1 kOe); these results are also shown in Figure 3b (dotted lines). The difference between the two cases of $R = 0.3$ and 1.75 is very large, which indicates that $H_{K,2}$ should be considered in the analysis. At $R = 1.75$, for example, the H_{ext} value for which the determinant is 0 is misallocated from 3.8 to 3.2 kOe when $H_{K,2}$ is disregarded; a new location that reveals a 0 value of the determinant emerges at $H_{\text{ext}} = 9.0$ kOe.

Figure 4a and b shows the results for G_x and G_y , which correspond to T_x and T_y in the conventional analysis at $R = 0.3$ and 1.75 , respectively. The results for the systems with $H_{K,2} = 0$ (solid lines)

Figure 3 Refined analytical results for $B_1^2 - A_1^2$ as a function of H_{ext} for systems with (a) $H_{K,2} = 0$ kOe and (b) $H_{K,2} = -1$ kOe. Two sets of results, for $R = 0.3$ (red lines) and $R = 1.75$ (blue lines), are shown. In **b**, the results calculated by disregarding $H_{K,2}$ (although it exists) are also shown (dotted lines).

Figure 4 Refined analytical results for G_x and G_y as functions of H_{ext} (a, b) and refined analytical results for ΔH_{DL} and ΔH_{FL} as functions of θ_M^o (c, d) for systems with $H_{K,2} = 0$ (solid lines) and $H_{K,2} = -1$ kOe (dashed lines). The left panels (a, c) and right panels (b, d) show the results for $R = 0.3$ and 1.75 , respectively. In **c**, **d**, the results calculated by disregarding $H_{K,2}$ (even though it exists) are also shown (dotted lines).

and $H_{K,2} = -1$ kOe (dashed lines) are shown. Note the H_{ext} values for which $G_x = G_y$ are 3.6 and 3.8 kOe for the systems with $H_{K,2} = 0$ kOe and $H_{K,2} = -1$ kOe, respectively. These H_{ext} values are identical to the H_{ext} values for which the determinant $B_1^2 - A_1^2 = 0$ (Figure 3a and b). This finding contradicts the case of the conventional analysis, where the H_{ext} value for which the determinant is 0 differs substantially from the H_{ext} value for which $T_x = T_y$ (Figures 1c and d, and 2d). With the new set of results for the determinant and the G ratios, the task of calculating the SO effective fields is straightforward; these results as a function of θ_M^o are shown in Figure 4c and d, for $R = 0.3$ and 1.75 , respectively. Two sets of results are shown: one set for the system with $H_{K,2} = 0$ kOe (solid lines) and one set for the system with $H_{K,2} = -1$ kOe (dashed lines). As shown in Figure 4c and d, in both systems, the calculated values of ΔH_{DL} and ΔH_{FL} are consistent with the input values for the macrospin simulation (over the entire θ_M^o range of $0^\circ \sim 82^\circ$); this finding demonstrates the reliability of the refined analysis. At $R = 0.3$, the agreement between the two systems is perfect to such an extent that the solid lines for the $H_{K,2} = 0$ kOe system completely overlap with the dashed lines for the $H_{K,2} = -1$ kOe system over the entire θ_M^o range. A similar behavior is observed at $R = 1.75$, with the difference that small peaks are observed at $\sim 43^\circ$ and

Figure 5 Contour plots that show deviation (in %) from input values of ΔH_{DL} (left panel) and ΔH_{FL} (right panel) as a function of θ_M^o and R . The results obtained from the conventional analytical method are shown in **a**, and the results obtained from the refined analytical method are shown in **b**. All results are obtained for the system with $H_{K,2} = -1$ kOe. In **a**, the regions filled with inclined lines indicate the θ_M^o range of no physical significance.

Figure 6 Results for ΔH_{DL} (a, c) and ΔH_{FL} (b, d) as functions of θ_M^o at three different I_0 values of 1.0 mA (black squares), 1.5 mA (red circles) and 2.0 mA (blue triangles), which are calculated using conventional (a, b) and refined (c, d) analytical methods. The results for ΔH_{DL} (e) and ΔH_{FL} (f) obtained at a θ_M^o value of 55° as functions of I_0 calculated using the conventional (black squares) and refined (red circles) analytical methods. Lines in **e**, **f** represent the least squares fit to the refined results.

$\sim 50^\circ$ for the systems with $H_{K,2} = 0$ kOe and $H_{K,2} = -1$ kOe, respectively, for which $B_1^2 - A_1^2 = 0$.

In systems with both $H_{K,1}^{\text{eff}}$ and $H_{K,2}$, the determinant $B_1^2 - A_1^2$ significantly differs if $H_{K,2}$ is disregarded (Figure 3b). A similar difference is expected for the calculated values of ΔH_{DL} and ΔH_{FL} (using Equation (27)), which are also shown in Figure 4c and d (dotted lines). For $R = 0.3$, the absolute values of ΔH_{DL} and ΔH_{FL} are underestimated in the θ_M^o range of $0^\circ \sim 60^\circ$ and overestimated in the

range of $60^\circ\sim 82^\circ$. This finding is reasonable based on the $H_{K,2}$ term, which is proportional to $\sin\theta_M^\circ\sin 3\theta_M^\circ$ (Equation (23)). For $R=1.75$, the differences are significant, with two divergences at $\sim 39^\circ$ and $\sim 80^\circ$ due to the misallocated H_{ext} fields of 3.2 and 9.0 kOe, for which $B_1^2 - A_1^2 = 0$ (Figure 3b). These results demonstrate that $H_{K,2}$ should not be neglected in the analysis of the harmonic measurement results in systems with both $H_{K,1}^{\text{eff}}$ and $H_{K,2}$.

Comparison of conventional and refined analyses over wide R range

Two typical R ratios of 0.3 and 1.75 have been considered. To test the refined analytical method over a wide R range, a systematic study was conducted by varying the R ratio from 0 to 2 in steps of 0.05 for the system with $H_{K,2} = -1$ kOe. Figure 5a and b displays contour plots that show the deviation (in %) from the input values of ΔH_{DL} (left panels) and the input values of ΔH_{FL} (right panels) as a function of θ_M° and R . The results calculated using the conventional analytical method are shown in Figure 5a, whereas the results calculated using the refined method are shown in Figure 5b. In the case of conventional solutions, the θ_M° range in which $H_{\text{ext}} > H_K^{\text{eff}}$ has no physical significance is indicated in Figure 5a as inclined lines. In Figure 5a and b, the solid lines indicate a deviation of 0.8% and the white regions indicate a minimum deviation of 4%. As shown in Figure 5a, the conventional solutions are valid over very limited ranges of θ_M° and R . For example, the R range in which the deviations are $< 4\%$ is 0.06–0.12 for ΔH_{DL} and 0.21–0.46 for ΔH_{FL} in the θ_M° range of $0^\circ\sim 52^\circ$. For R values higher than 1.1, the validity range is even more limited for both ΔH_{DL} and ΔH_{FL} ; specifically, the θ_M° values for which the deviations are $< 4\%$ are 4.5° at $R=1.1$ and 7.9° at $R=2.0$ for ΔH_{DL} and 4.5° at $R=1.1$ and 9.4° at $R=2.0$ for ΔH_{FL} . In the intermediate R range of 0.9–1.1, the deviations are always larger than 4%. The accuracy of the calculated results improves significantly with the use of the refined method, as shown in Figure 5b. With the z component of H_{ext} considered in the refined analysis, no region has physical insignificance. The predictions made in the refined analysis are highly accurate. For $R < 0.85$, the deviations are $< 0.4\%$ over the entire θ_M° range of $0^\circ\sim 82^\circ$ for both ΔH_{DL} and ΔH_{FL} . Even for $R > 0.85$, the deviations are $< 0.8\%$ for both ΔH_{DL} and ΔH_{FL} over the entire region, except in the regions marked by solid lines, where the deviations are large due to the existence of divergences (zero determinants).

Analysis of experimental results

An additional test of the refined method was performed by analyzing the experimental results of the harmonic Hall voltage measurements for a stack with the following structure: Si substrate (wet-oxidized)/Ta (5 nm)/Pt (5 nm)/Co (0.6 nm)/MgO (2 nm)/Ta (2 nm). Refer to Supplementary Figures S2 and S3 for Hall bar dimensions. The results were obtained at three different I_0 values: 1.0; 1.5; and 2.0 mA. The magnetization direction was controlled by H_{ext} , which was swept from +90 to -90 kOe with two different directions: $\theta_H = 85^\circ$ and $\varphi_H = 0^\circ$; and $\theta_H = 85^\circ$ and $\varphi_H = 90^\circ$. The values of $H_{K,1}^{\text{eff}}$ and $H_{K,2}$, which were extracted using the Generalized Sucksmith–Thompson method,³⁵ were 33.1 and -8.1 kOe, respectively. The R ratio of the sample was measured to be 0.423. Figure 6a–d shows the results for ΔH_{DL} (a and c) and ΔH_{FL} (b and d) as functions of θ_M° at three different I_0 values: 1.0 mA (black squares); 1.5 mA (red circles); and 2.0 mA (blue triangles). Both conventional (a and b) and refined analytical methods (c and d) were used to analyze the experimental results. The results from the conventional method show incorrect divergences at $\theta_M^\circ \sim 60^\circ$, whereas the results from the refined method do not show this behavior over the entire θ_M° range. Note that both ΔH_{DL}

and ΔH_{FL} depend on θ_M° . The values of ΔH_{DL} and ΔH_{FL} should be proportional to I_0 , with zero values at $I_0 = 0$.¹⁵ The expectation is only satisfied for the results extracted using the refined method, as shown in Figure 6e and f, where the results for ΔH_{DL} (red circles) and ΔH_{FL} (black squares) obtained at a fixed θ_M° value of 55° are shown as functions of I_0 . A large deviation from the linearity is particularly noted for the ΔH_{FL} results calculated using the conventional method. Refer to Supplementary Figures S4–S6 for detailed results.

DISCUSSION

The test of the conventional analytical method, which involves an analysis of the macrospin simulation results, indicates that its validity range is very limited in terms of θ_M° and R , which is primarily attributed to the singularities involved in Cramer's rule at incorrect θ_M° values. This problem is overcome by the refined analytical method proposed in this study with detailed analytical equations, in which both the z component of H_{ext} and the second-order PMA are considered. The SO effective fields that are extracted using the refined analytical method are consistent with the input SO effective fields that are used for the macrospin simulation over the entire θ_M° range and over a wide R range of zero to two. For $R < 0.85$, deviations from the input SO effective fields are $< 0.4\%$ over the entire θ_M° range of $0^\circ\sim 82^\circ$ for both ΔH_{DL} and ΔH_{FL} . Even at $R > 0.85$, the deviations are $< 0.8\%$ for both ΔH_{DL} and ΔH_{FL} over the entire region, with the exception in some limited regions that exhibit singularities. The accuracy of the refined method is reconfirmed by an additional comparative study that involves analyzing the experimental results of harmonic Hall voltage measurements for a Pt/Co/MgO structure. An accurate analysis of the harmonic Hall voltage measurement results by the refined analytical method over very wide ranges of θ_M° and R will significantly contribute to the identification of a dominant mechanism of the SOT and the development of highly efficient SOT devices.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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