Spin-orbit torque in a bulk perpendicular magnetic anisotropy Pd/FePd/MgO system

Hwang-Rae Lee, Kyujoon Lee, Jaehun Cho, Young-Ha Choi, Chun-Yeol You, Myung-Hwa Jung, Frédéric Bonell, Yoichi Shiota, Shinji Miwa & Yoshishige Suzuki

1Department of Physics, Inha University, Incheon 402-751, Korea, 2Department of Physics, Sogang University, Seoul 121-742, Korea, 3Graduate School of Engineering Science, Osaka University, Osaka 560-8531, Japan, 4CREST, Japan Science Technology Agency, Saitama 332-0012, Japan.

Spin-orbit torques, including the Rashba and spin Hall effects, have been widely observed and investigated in various systems. Since interesting spin-orbit torque (SOT) arises at the interface between heavy nonmagnetic metals and ferromagnetic metals, most studies have focused on the ultra-thin ferromagnetic layer with interface perpendicular magnetic anisotropy. Here, we measured the effective longitudinal and transverse fields of bulk perpendicular magnetic anisotropy Pd/FePd (1.54 to 2.43 nm)/MgO systems using harmonic methods with careful correction procedures. We found that in our range of thicknesses, the effective longitudinal and transverse fields are five to ten times larger than those reported in interface perpendicular magnetic anisotropy systems. The observed magnitude and thickness dependence of the effective fields suggest that the SOT do not have a purely interfacial origin in our samples.

Since the discovery of spin transfer torque (STT), the method of manipulation of the magnetization direction has been shifted from the Oersted field to the spin current1,2. STT has been widely studied, and it is a building block of next-generation non-volatile random access memory. More recently, another magnetization manipulation method has been reported for a few atomic magnetic layers sandwiched with heavy metal and oxide layers3–8. The charge current passing through a heavy nonmagnetic metal layer produces effective fields by two known mechanisms: the Rashba effect and/or the spin Hall effect (SHE). Both phenomena are related to strong spin-orbit coupling, so they are called the spin-orbit torque (SOT) effect. It has been reported that SOT can alter the spin dynamics in an ultra-thin ferromagnetic layer through the effective fields9, and move the domain wall much faster than by STT only. Also, the domain wall motion direction can be opposite to the electron flow10. At the early stage of the study, it has been known that the Rashba effect generates a transverse effective field (or field-like term), while the SHE produces a longitudinal effective field (or damping-like term). However, refined models recently showed that both mechanisms generate both transverse and longitudinal effective fields11; thus, distinguishing between them remains challenging issues. Using SOT combines several advantages compared to the STT in spin valves. For example, the SHE uses only pure spin current that is generated in nonmagnetic heavy metals to manipulate the magnetization of the magnetic layer, and requires only a single magnetic layer. In contrast, a second reference ferromagnetic layer is inevitable in the STT technique, and large currents have to be set across a tunnel junction. The efficiency of the SOT is superior to that of the STT.

It is believed that the Rashba effect is generated in the conduction electrons at the interface between the nonmagnetic heavy metal and the magnetic layer by strong spin-orbit coupling. Conduction electrons under the finite potential gradient feel the effective magnetic field due to the strong spin-orbit coupling, as described by special relativity. And, the conduction electrons are strongly coupled with the localized d-electrons. Therefore, the spin motions of the localized d-electrons are also affected by the Rashba effective field12. On the other hand, the SHE is a diffusive spin current from the adjacent heavy nonmagnetic metal volume to the magnetic layer. The injected spin currents rapidly decay due to Fermi surface averaging, similar to STT in a metallic spin valve structure13–16. Therefore, both SOTs can be handled as interface effective fields.

Most studies have been performed for only a few atomic magnetic layers having interfacial perpendicular magnetic anisotropy (PMA), such as Co or CoFeB, which are sandwiched between nonmagnetic heavy metals (Pt, Ta, W) and oxide layers (AlOx, MgO). The interfacial PMA system maintains its out-of-plane easy axis only for limited thickness ranges. For example, Kim et al. investigated a Ta/CoFeB/MgO system with a 0.9 to 1.3 nm-thick...
CoFeB layer. Due to the interfacial nature of PMA, it shows an inverse proportionality to the ferromagnetic layer thickness. Fan et al. studied in-plane anisotropy materials of Py (10 nm thickness) by measuring transport properties, and the crossover from the perpendicular anisotropy of thin CoFeB to the in-plane anisotropy of thick CoFeB, by measuring the magneto-optical Kerr effect. They found that the longitudinal effective field is inversely proportional to the thickness of the magnetic layer, and that the transverse effective field decreases rapidly when the magnetic layer is thicker than 1 nm. Jamali et al. investigated a [Co (0.2 nm)/Pd (0.7 nm)] multilayer system and reported huge effective fields for the longitudinal (11.7 Oe/10¹⁰ A/m²) and transverse (50.25 Oe/10¹⁰ A/m²) directions. This suggests that the Rashba effective field is important despite the symmetry structure of their samples. More recently, Qiu et al. reported temperature and angular dependences of SOT in a Ta/CoFeB/MgO system. They found that the transverse field decreased with decreasing temperature, whereas the longitudinal field showed weak temperature dependence. This implies that the physical origins of two effective fields may differ. So far, reported results show that the purely interfacial nature of SOT is questionable.

In this study, we examined SOT in a Pd/FePd (1.54 to 2.43 nm)/MgO system with a bulk PMA due to the L₁₀ crystalline structure of FePd. By virtue of the bulk L₁₀ crystalline PMA, we could extend the SOT research to much thicker ferromagnetic layer systems compared to other groups. With careful analysis by considering the planar Hall effect and Oersted field contributions, we found that the longitudinal effective field is five to ten times stronger than, or comparable to, that of a Ta/CoFeB/MgO when we considered the ferromagnetic layer thickness. The experimentally observed thickness dependencies of the transverse and longitudinal effective fields arise a question about the interface origin of SOT.

Results
Measurement of the longitudinal and transverse effective fields. Since the origins of the effective transverse and longitudinal fields are not easily distinguishable, we focused on systematic measurements of the effective longitudinal and transverse fields in Pd/L₁₀-FePd (1.54 to 2.43 nm)/MgO wedge samples, as shown in Fig. 1a. In this geometry, the applied current produces two effects. SHE separates the charge current to up- and down-spin current, as shown in Fig. 1b, and it mainly leads to the longitudinal effective field (Hₗ). The potential gradient at the interface generates the Rashba effect, which causes the transverse effective field (Hₜ), as shown in Fig. 1b. However, the Rashba effect (SHE) can also generate the longitudinal (transverse) fields.

The measurement geometry for the 2θ method with the coordinate system is shown in Figs. 1c and d. A 100 μm × 10 μm wire was connected to the two electric pads with two pairs of Hall pads. The measured Hall signals are comprised of the anomalous Hall effect (AHE) and the planar Hall effect (PHE).

\[ V_{\text{Hall}} = \frac{V_{\text{AHE}}}{2} \cos \theta + \frac{V_{\text{PHE}}}{2} \sin^2 \theta \sin 2\phi. \]

Here, \( V_{\text{AHE}} = I_{\text{P}}R_{\text{AHE}} \) and \( V_{\text{PHE}} = I_{\text{P}}R_{\text{PHE}} \) are the Hall voltages of the AHE and PHE and \( R_{\text{AHE}} \) and \( R_{\text{PHE}} \) are corresponding Hall resistances, respectively. The applied alternating current (AC) current is given by \( I(t) = I_{\text{dc}} \sin \omega t \). The polar angle \( \theta \) and azimuthal angle \( \phi \) of the magnetization direction are defined in the inset of Fig. 1b, and they are determined mainly by the external magnetic field and the system anisotropy energy. However, they oscillate with a modulation frequency of \( \omega \) due to the external AC effective field. Therefore, the measured Hall voltage can be expressed as:

\[ V_{\text{Hall}} = V_{\text{dc}} + V_{\text{ac}} \sin \omega t + V_{\text{2ac}} \cos 2\omega t + \ldots. \]

It is known that the effective longitudinal (\( H_{\text{L},y}-\text{axis} \)) and transverse (\( H_{\text{T}},x\)-axis) fields can be determined from the ratio between the slope of the out-of-phase second harmonics of the Hall voltage and the curvature of the in-phase first harmonics, as follows:

\[ H_{\text{LT}} = \frac{-2}{\psi_{\text{L}}^{\text{ext},y}} \frac{\partial^2 V_{\text{2ac}}}{\partial H_{\text{ext},y}^2}; \]

where \( H_{\text{ext},y} \) are the external applied transverse (\( x \)) and longitudinal (\( y \)) fields during the AHE measurements, and \( V_{\text{dc}} \) and \( V_{\text{ac}} \) are the first and second harmonics of the AHE voltages, respectively. Equation (3) is valid only when the PHE contribution is ignored. However, the PHE cannot be generally ignored and must be carefully considered. With valid approximations, we obtained the following equations (see Sec. S2 in Supplementary Information):

\[ H_{\text{L},\text{meas}} = \frac{-2}{\psi_{\text{L}}^{\text{ext},y}} \frac{\partial^2 V_{\text{2ac}}}{\partial H_{\text{ext},y}^2} \pm (H_L - 2\chi H_T), \]

\[ H_{\text{T},\text{meas}} = \frac{-2}{\psi_{\text{T}}^{\text{ext},x}} \frac{\partial^2 V_{\text{2ac}}}{\partial H_{\text{ext},x}^2} \pm (H_T - 2\chi H_L). \]

where \( \chi = R_{\text{PHE}}/R_{\text{AHE}} \) is the resistance ratio between AHE and PHE. \( H_{\text{L},\text{meas}} \) and \( H_{\text{T},\text{meas}} \) are the experimentally measured results from the first and second harmonic signals, respectively, and the ± sign represents up and down magnetization (±Mₛ), respectively. \( H_L \) and \( H_T \) represent the real effective field that we want to determine in the present experiments. Equations (4) and (5) describe the effective fields obtained by Kim et al. and they are actually mixed signals of the longitudinal and transverse effective fields whenever the PHE is nonzero. From Eqs. (4) and (5), \( H_L \) and \( H_T \) can be readily extracted if \( \chi \) is known, as follows:

\[ H_L = \frac{H_{\text{L},\text{meas}} \pm 2H_{\text{T},\text{meas}}}{1 - 4\chi^2}, \]

\[ H_T = \frac{H_{\text{T},\text{meas}} \pm 2H_{\text{L},\text{meas}}}{1 - 4\chi^2}, \]

where the ± sign represents the up and down magnetizations. With corrected expressions, one can separate the PHE contribution in the measurement signals. We note that if \( \chi \) is small, the PHE contribution is negligible. More details are given in the Supplementary Information (S2).

Experimental results, and other contributions to the longitudinal and transverse effective fields. Figures 2a–f show typical experimental results in the present study for a 1.54 nm FePd sample. Figure 2a shows an AHE hysteresis loop. The Hall voltage was measured with the external field in the z-direction (\( H_z \)), and the detecting current was small enough so that the effective field due to the SOT is negligible. Figure 2b shows the first harmonics of Hall voltage \( V_{\text{dc}} \) with the transverse and longitudinal external fields (\( H_x \) and \( H_y \)). With moderate field strength, the first harmonic signals are approximated by a parabola to the external field, as shown in Fig. 2b. The small difference in curvatures between the transverse and longitudinal \( V_{\text{dc}} \) are due to the finite in-plane shape anisotropy. We applied a moderate strength field to keep \( \theta \approx 1 \). More details regarding \( V_{\text{dc}} \) are given in Supplementary Information (S2). Figures 2d and e are the out-of-plane transverse and longitudinal \( V_{\text{2ac}} \) for the up and down magnetizations with corresponding external in-plane fields. Here, the transverse (longitudinal) \( V_{\text{2ac}} \) is odd (even) with respect to the magnetization inversion due to the symmetry of the given problem. Note that the \( V_{\text{2ac}} \) signals are much more scattered than those reported by other groups. The main reason for the scattered signals is the 18.5 nm-thick Pd underlayer, which is necessary to promote better L₁₀ crystal structure of the FePd layer. A large shunt current flowed through the thick Pd underlayer, and only a small part of the current contributed to the AHE signal.
Despite the scattered signals, it is sufficient to determine the slope of $V_{2v}$, as shown in Figs. 2d and e.

The PHE measurement is shown in Fig. 2c. In order to measure the PHE, we applied a $1.0$ kOe longitudinal field (that is the maximum field of our apparatus). It is sufficient to lay the magnetization down on the in-plane for the thinnest FePd (1.54 nm) sample, whose perpendicular magnetic anisotropy is the smallest. While keeping the $1.0$ kOe longitudinal field, we added a small transverse field ($\sim 100$ Oe) to detect the planar Hall signal. Since the in-plane anisotropy field $H_{in} = (N_x - N_y)M_s \sim 100$ Oe was much smaller than the external longitudinal field ($\pm 1.0$ kOe), we ignored the effect of the in-plane anisotropy, and the azimuthal angle $\phi$ of the magnetization is almost the same as the in-plane field angle $\phi_H = \pi/2 - \arctan(H_x/H_y)$. With these assumptions, we extracted $V_{PHE}$ from Eq. (1), and then determined $V_{PHE} = 6.01 \pm 0.05$ $\mu$V and $V_{AHE} = 25.4$ $\mu$V from Figs. 2a and c. This means that the PHE contribution, $\chi = 0.237$, is not negligible in the present experiments. Thus, we used $\chi$ for a careful analysis of our experimental data using Eqs. (6) and (7).

Another important contribution in the measured Hall voltages is the anomalous Nernst-Ettingshausen (ANE) effect due to inhomogeneous heating across the Hall bar geometry. This inhomogeneous heat distribution is unavoidable in the harmonic measurement experiments. The ANE contribution is described by Garello et al. in the Supplementary Information (S4). In summary, the Joule heating due to the applied current is about 40 to 50 K for a maximum current density of $2.7 \times 10^{11}$ A/m$^2$. If the temperature increment alters the magnetic properties, it is clear that the behavior of the AHE with respect to the applied voltage must deviate from the linear. In addition, the curvature of $V_{2v}$ data is related to the effective perpendicular magnetic anisotropy energy, must be changed with the applied voltage. However, the AHE signal of the thinnest FePd is linear with respect to the applied AC voltage until reaching the maximum applied current density (Fig. S5a in the Supplementary Information), and the $V_{2v}$ data with various voltages collapse to the single curve (Fig. S5b).

These results indicate that the magnetic properties of the sample are not noticeably changed by the Joule heating despite the temper-
nature increment of \(-50\) K. Based on these observations, we suggest that the Joule heating effect is not important in the present experiments.

Finally, the Oersted field contribution is also important in our sample structure. Since the nonmagnetic layer composed of 1.5 nm-thick Cr and 18.5 nm-thick Pd is thicker than that in the structures of other groups, the total current is larger for a given current density. The estimated Oersted field is

\[ H_{oe} = J_{Pd} \frac{1}{2} \frac{1}{1 + 2x^2} \]

for 10^{10} A/m^2. This value is comparable to the strength for the effective transverse field, and is almost ten times larger than that reported by Kim et al.7 It is also due to the thicker Pd underlayer. Importantly, the Oersted field is positive in our experimental geometry, while the measured transverse effective field is always negative. This means that the Oersted field always reduces the magnitude of the transverse effective field due to the SOT. We will correct the Oersted field contribution in the final results.

Thickness dependence of the SOT effective fields. Figure 3a shows the measured longitudinal and transverse effective fields (\(-H_{L,\text{measured}}\) and \(H_{T,\text{measured}}\)) determined using Eqs. (4) and (5), respectively, which are mixtures of the pure longitudinal and transverse effective fields. We plot \(-H_{L,\text{measured}}\) and \(H_{T,\text{measured}}\) in Fig. 3a for \(t_{FePd} = 1.54, 1.73, 2.13,\) and 2.43 nm, together with linear fits. The applied AC voltage was converted to the corresponding current density \(J\). As previously discussed, the effective field does not deviate from linear behavior, which implies that Joule heating is not significant in our experiments. We note that the signs of \(H_L\) are negative (positive) for up (down) magnetizations, and the signs of \(H_T\) are always negative for up and down magnetizations (Figs. 2d and e, and Eqs. (4) and (5)). Therefore, we plotted \((H_{L,+} - H_{L,-})/2\) and \((H_{T,+} + H_{T,-})/2\) in Fig. 3a for easy comparisons. With these measured effective fields and the ratio of PHE and AHE (\(x = 0.237\)), we corrected the effective fields using Eqs. (6) and (7). Due to the small value of \(1/(1 - 4x^2) = 1.29\) in Eqs. (6) and (7), and mixed contributions of the pure longitudinal and transverse fields, the corrected effective fields are somewhat larger than the measured ones. Especially, the corrected transverse effective fields are much larger than the measured fields. For example, the measured transverse fields of 1.54 nm are the smallest among the series (Fig. 3a), but they turn to be the largest after correction (Fig. 3b).

The corrected transverse fields in Fig. 3 still include the Oersted field. Therefore, an additional conversion, \(H_{T,\text{SOT}} = H_{T,\text{corrected}} - \)
H_{\text{measured}}$ is required to obtain the pure effective field, again with the proper sign considerations ($H_{L,\text{measured}} < 0$ and $H_{T,\text{measured}} > 0$). We compare these fully corrected effective fields with the results of other groups in Fig. 4. We scaled the effective fields from the literatures\textsuperscript{7,9,18,20,24} to a current density of $10^{10}$ A/m$^2$ (assuming linearity with current), and plotted them as a function of the inverse ferromagnetic layer thickness, $1/t_{\text{FM}}$. For easy comparison, we took the absolute values of each effective field. Qiu \textit{et al.}\textsuperscript{20} reported the angular and temperature dependences of the effective fields in the Ta(2)/CoFeB(0.8)/MgO(2)/SiO$_2$(3); we took only the value at 300 K and a polar angle of $4^\circ$, which are conditions similar to ours. They reported $24.4$ and $19.42$ Oe/10$^{10}$ A/m$^2$ for the longitudinal and transverse directions, respectively. Even though the thickness of their CoFeB layer is much thinner than the present study, the longitudinal field is comparable to ours, and the transverse field is larger.

Despite their remarkable works, we did not take data from the works of Fan \textit{et al.}\textsuperscript{17} and Jamali \textit{et al.}\textsuperscript{19} in Fig. 4. Fan \textit{et al.}\textsuperscript{17} investigated thick Py ($\geq 2$ nm), which is out of the range of our plot. In addition, the effective field is relatively small ($\leq 1$ Oe/10$^{10}$ A/m$^2$) for longitudinal and transverse fields. Jamali \textit{et al.}\textsuperscript{19} studied a Co (0.2 nm)/Pd (0.7 nm) multilayer system and found huge longitudinal (11.7 Oe/10$^{10}$ A/m$^2$) and transverse (50.25 Oe/10$^{10}$ A/m$^2$) fields. The reason for the huge effective field in a 19.8 nm-thick [Co/Pd]$_{22}$ multilayer system is not clear. Therefore, we should omit these data sets for better readability of Fig. 4.

As shown in Fig. 4, the effective longitudinal field in the present work exhibits meaningful $1/t_{\text{FM}}$ dependence, whereas the transverse field is almost independent of $t_{\text{FM}}$. Due to the limited number of data points ($4 t_{\text{FM}}$) and scattered data points, it is not quite clear why the longitudinal field is proportional to $1/t_{\text{FM}}$ in the present study. The inverse proportionality to $t_{\text{FM}}$ means that the SOT effective field is purely an interface effect. Based on this argument, both effective fields must be proportional to $1/t_{\text{FM}}$ and pass through the origin. Such linear dependence is observed in the results of Fan \textit{et al.}\textsuperscript{18} for a wide range of Ti/CoFeB/Pt thicknesses across the perpendicular to in-plane anisotropy regions for the longitudinal region, as shown in Fig. 4 (the blue closed circles represent $H_L$). However, the linearity of the transverse fields is not very clear (the blue open circles represent $H_T$). Kim \textit{et al.}\textsuperscript{7} also reported that the transverse fields show linear dependence (green open triangles with a straight line), but the longitudinal field is not linear in the Ta/CoFeB/MgO system. However, their results have a nonzero crossing point of $1/t_{\text{FM}}$, which is physically unreasonable. One possible reason is that Kim \textit{et al.}\textsuperscript{7} did not take into account the PHE contribution properly in their analysis. Quite generally, these deviations from a $1/t_{\text{FM}}$ dependence make the interfacial nature of the SOT questionable. In our case, the longit-

Figure 3 | Longitudinal and transverse effective fields. (a) The measured effective fields, determined using Eqs. (4) and (5) for 1.54 to 2.43 nm-thick FePd. The signs of $H_{L,\text{measured}}$ and $H_{T,\text{measured}}$ are negative, and the solid line represents linear fittings. (b) Corrected effective fields based on Eqs. (6) and (7) with $\chi = 0.237$. 

![Figure 3](image-url)
We now discuss possible origins of the relatively large SOT in our multilayered system, and ascribed the large SOT to the large asymmetry (see S6 in supplementary information). This means that there is certain thickness (about 3 atomic layers) to be stabilized in the film, and the anisotropy is found to increase rapidly with the FePd thickness.

The ideal FePd L1₀ structure shows inversion symmetry, so that only the potential gradient at the very Pd/FePd interface should be source of Rashba effect. However, the large transverse field observed (given the weak spin-orbit coupling in Pd compared to Pt or Ta), and the weak thickness dependence of it suggest some deviation from a purely interfacial Rashba effect. Actually, the L1₀ structure requires a certain thickness (about 3 atomic layers) to be stabilized in the film, and the anisotropy is found to increase rapidly with the FePd thickness (see S6 in supplementary information). This means that there is an extended gradient of chemical ordering near the Pd/FePd interface, which could be a source of potential gradient inside the FePd.

In summary, we observed large longitudinal and transverse effective fields due to the SOT in a bulk PMA Pd/FePd/MgO system. The pure longitudinal and transverse effective fields were extracted with a careful data analysis taking into account the finite PHE. The observed magnitude and thickness dependence of the effective fields suggest that the SOT do not have a purely interfacial origin.
Methods
Sample preparations and micro-fabrication. The films were grown using molecular beam epitaxy. The stacking consisted of a MgO (7)/Cr (1.5)/Pd (18.5)/FeCo/Pd (15) multilayer deposited on a single-crystal MgO (001) substrate (thicknesses in nanometers). The FePd wedge layer thickness, $t_{\text{FePd}}$, was varied from 1.54 to 2.43 nm. The Pd buffer was annealed for 20 min at 400 °C to obtain a flat surface. Wedge-shaped FePd was grown by co-deposition of Fe and Pd sources with a substrate temperature of 350 °C to induce L1$_2$ ordering during growth with the c axis normal to the plane. All layers except the FePd were deposited at room temperature. More details of the structure are explained in the previous report and Supplementary Information S1. The film was patterned into a hall bar using electron beam lithography and an Ar ion milling technique.

Measurement method. All the measurements were executed in a three-dimensional magnet probe station, where the magnetic field could be changed in any random direction without rotating the device. We used the harmonics technique to measure the current-induced effective field. The Hall voltage was measured by modulating an AC current at 307.3 Hz with two lock-in amplifiers: one for the in-phase first harmonics and the other for the out-of-phase (90° off) second harmonics. We swept the magnetic field along the x and y directions to obtain the transverse and longitudinal effective fields, respectively.

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Author contributions
H.-R.L., K.L. and Y.-H.C. carried out the harmonics measurements. M.-H.J. supervised the measurements. J.C. and C.-Y.Y. analyzed the experimental data. F.R. and Y.Sb. prepared the samples. S.M. and Y.Su. supervised the sample preparations. The manuscript was prepared and the project was designed by M.-H.J., C.-Y.Y. and Y.Su. H.-R.L. and K.L. contributed as first authors. Correspondence and requests for materials should be addressed to C.-Y.Y. or M.-H.J.

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