Research article

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Interface-induced field-like optical spin torque in a ferromagnet/heavy metal heterostructure

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Abstract: The manipulation of magnetization in a metallic ferromagnet by using optical helicity has been much attracted attention for future opto-spintronic devices. The optical helicity-induced torques on the magnetization, *optical spin torque*, have been observed in ferromagnetic thin films recently. However, the interfacial effect of the optical spin torque in ferromagnet/nonmagnetic heavy metal heterostructures have not been addressed so far, which are widely utilized to efficiently control magnetization via electrical means. Here, we studied optical spin torque vectors in the ferromagnet/nonmagnetic heavy metal heterostructures and observed that in-plane field-like optical spin torque was significantly increased with decreasing ferromagnetic layer thicknesses. The interfacial field-like optical spin torque was explained by the optical Rashba–Edelstein effect caused by the structural inversion symmetry breaking. This work will aid in the efficient optical manipulation of thin film nanomagnets using optical helicity.

Keywords: optical helicity; optical spin; optical spin torque; photospintronics; spin–orbit torque.

1 Introduction

Recently, ferromagnetic/nonmagnetic heavy metal heterostructures have been widely used for manipulating magnetization via electrical means in the field of spin-orbitronics [1]. The spin Hall effect and/or the Rashba–Edelstein effect have been observed in heterostructures, which enable the efficient control of thin film magnets [2, 3]. A previous study reported that the magnetization of an ultrathin Co film sandwiched between Pt and AlO_x layers can be switched by in-plane current injection. This is attributed to the torque generated by the Rashba field due to the asymmetric heterostructure [2]. Further, it was demonstrated that the spin-transfer torque in a ferromagnetic layer can be induced by the spin Hall effect in adjacent nonmagnetic heavy metal layers such as Ta and Pt [3]. Both these torques, which are generally called spin–orbit torques, can be considered to be caused by the interface effect of the heterostructure and the bulk effect of the nonmagnetic layer [4, 5].

Analogous to the electrical generation of the spin–orbit torque, the optical manipulation of magnetization by using the spin-angular momentum of light, namely optical helicity, in metallic thin films has been investigated extensively in experimental [6–10] and theoretical studies [11–13]. This optical manipulation will have future applications in ultrafast and low energy consumption opto-spintronic devices [14, 15]. Although the transfer of spin-angular momentum from light to thin film metallic magnets has been considered to be marginal, accumulative magnetization switching induced by optical helicity has been observed [6–10], which stems from magnetic circular dichroism [16, 17]. In contrast to the accumulative magnetization switching induced by magnetic circular dichroism, a recent experimental study observed that magnetization reversal is also induced by a single circularly polarized laser pulse [10]. In addition, magnetization precession dynamics induced by optical helicity in a metallic thin film magnet was observed [18, 19]. The torque on magnetization induced by optical helicity in metallic heterostructures can generate a helicity-dependent terahertz emission [20, 21], in which the terahertz photocurrent is produced due to the inversion symmetry breaking of

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the thin film structure. These experiments indicated that the optical helicity-induced torque, which we term *optical spin torque*, has a certain effect on metallic thin film magnets, although many previous studies have used magnetic semiconductors or insulators to observe the effect of optical helicity [22–29]. Optical spin-transfer torque as well as optical spin-orbit torque have been observed in a magnetic semiconductor GaMnAs, respectively in a study by Němec et al. [27, 28], the mechanism of the torque is different with the case of the metal.

In the initial observations of optical spin torque in metallic thin film magnets, optical helicity-induced magnetization precession was mainly explained by the inverse Faraday effect in ferromagnetic metals [18], in which the optical spin torque has a field-like form, $\mathbf{m} \times \mathbf{s}$, where $\mathbf{m}$ and $\mathbf{s}$ are the magnetization and spin direction pointing along the wave vector of the light, i.e., $\mathbf{s} \propto \mathbf{E} \times \mathbf{E}^*$. However, the recent observations of the optical helicity-induced magnetization precession in Co/Pt bilayer can be explained using spin-transfer mechanism as follows [19]. The spin $\mathbf{s}$ generated by the optical helicity in the Pt layer was due to the optical orientation, and it transfers its angular momentum to an adjacent Co layer via the spin-transfer torque effect. It was concluded that the magnetization precession in Co was excited by the out-of-plane

spin-transfer torque in the form $\mathbf{m} \times (\mathbf{m} \times \mathbf{s})$ as shown in Figure 1(a).

The study by Choi et al. [19] have studied the effect of circularly polarized light with different thickness of Co and Pt. Then, they discussed the thickness dependences in terms of light absorption and optical orientation in nonmagnetic heavy metal Pt. However, such spin-orbit induced optical effect can also arise from interfaces, like optical Rashba–Edelstein effect proposed in the past [30]. In this article, we experimentally study the optical spin torque for magnetic and heavy metal heterostructures in detail and report, for the first time, a new type of *interface-induced* field-like optical spin torque, as schematically shown in Figure 1(b).

2 Experimental procedure

Thin film samples were prepared by the magnetron sputtering method. MgO (10)/Fe₅₀Co₅₀ (d_{FeCo})/Pt (d_{Pt}) (thickness is in nm) films were deposited on thermally oxidized Si/SiO₂ substrate. To measure circularly polarized laser pulse induced magnetization dynamics, the conventional time-resolved magneto-optical Kerr effect (MOKE) setup was employed [31, 32]. The wavelength, pulse duration, and repetition rate of the pulse laser used in this study were 800 nm, ~120 fs, and 1 kHz, respectively. The pump pulse was irradiated on the film at an angle of 10° measured from the film normal. The polar MOKE of the

Figure 1: (a) Schematic illustration of optical spin torque induced by the optical orientation in the heavy metal layer. (b) Field-like optical spin torque induced by the circularly polarized laser due to interface spin-orbit coupling. (c) Typical magnetization precessional dynamics excited by circularly polarized laser pulse with different optical helicity for 2-nm-thick FeCo/3-nm-thick Pt thin film.

probe laser pulse was detected to measure the magnetization component normal to the film plane. To detect the pump pulse induced change in the polar MOKE signal, the pump pulse was modulated with a frequency of 360 Hz by using a mechanical chopper. The pump induced change in the MOKE signal was detected by a lock-in amplifier. The pump fluence F_p used in this study was fixed to 8.3 J/m^2 . An in-plane external magnetic field up to 2 T was applied.

3 Results and discussions

3.1 Optical helicity-induced magnetization dynamics and its magnetic field dependence

Figure 1(c) shows typical femtosecond laser pulse induced magnetization dynamics where left circularly polarized (LCP) laser and right circularly polarized (RCP) laser were irradiated and a 2 T in-plane magnetic field was applied. A change in the initial phase of magnetization precession was observed when light of different optical helicity was irradiated on the film. A slight difference was observed in precessional amplitude owing to sample misalignment. This is because thermal demagnetization can induce magnetization precession if the magnetic field is slightly tilted toward the out-of-plane direction. The signal at approximately $t = 0$ could be due to the fact that MOKE senses the orbital magnetic moment excited by the optical helicity pulse, the so-called specular inverse Faraday effect and specular inverse optical Kerr effect [33–36]. Figure 2(a) shows the magnetic field dependence of the optical helicity-induced magnetization precession in the MgO/FeCo(2)/Pt(3) thin film. The magnetization precession dynamics after the initial peak were fitted using a sinusoidal decay function as $m_0 \exp(-t/\tau) \sin(2\pi f t + \phi_0)$, where m_0, f, τ, ϕ_0 are amplitude, frequency, lifetime, and initial phase of magnetization precession, respectively. The precession frequency increases with an increase in the magnetic field, while the amplitude does not significantly depend on the magnetic field. The precession frequency f is evaluated by the fitting plotted as a function of the magnetic field $\mu_0 H$ in Figure 2(b). The f vs $\mu_0 H$ curve is fitted by the Kittel formula as, $f = \gamma \sqrt{\mu_0 H (\mu_0 H + \mu_0 M_{\text{eff}})} / (2\pi)$, where γ and $\mu_0 M_{\text{eff}}$ are gyromagnetic ratio and an effective demagnetization field. Figure 2(c) shows the inverse lifetime $1/\tau$ evaluated by the fitting plotted as a function of $\mu_0 H$. The $1/\tau$ vs $\mu_0 H$ plot is well explained by the theoretical relation $1/\tau = \alpha \gamma (\mu_0 H + \mu_0 M_{\text{eff}}/2)$, where the Gilbert damping parameter α is 0.019. The large value of α compared with α for bulk FeCo (~ 0.002 [37]) is due to the spin-pumping effect at the FeCo/Pt interface [38]. Figure 2(d) shows the averaged

precession amplitude ($(|m_0(\text{LCP})| + |m_0(\text{RCP})|)/2$) plotted as a function of precession frequency. The precession frequency does not have a significant influence on the precession amplitude, indicating non-thermal ultrafast impulsive excitation induced by the optical spin torque.

3.2 Thickness dependences of the optical helicity-induced magnetization precession

To understand the excitation mechanism of the optical helicity-induced magnetization precession in FeCo/Pt bilayer, nonmagnetic Pt and ferromagnetic FeCo thickness dependence were measured as shown in Figure 3. To exclude the effect of the sample misalignment as mentioned above, we measured the signal for both +2 T and -2 T, and the average was obtained. In addition, the MOKE signal was normalized by the static MOKE voltage to detect the change in normalized magnetization. Figure 3(a) and (b) show the Pt thickness d_{Pt} and FeCo thickness d_{FeCo} dependences of the optical helicity-induced magnetization precession. It was found that the precession amplitude initially increased with increasing d_{Pt} from 2 to 5 nm and then slightly decreased at $d_{\text{Pt}} = 10$ nm. The decrease in amplitude with increasing d_{FeCo} [Figure 3(b)] can be

Figure 2: (a) Circularly polarized laser pulse induced magnetization precession in 2-nm-thick FeCo/3-nm-thick Pt bilayer with different magnetic fields $\mu_0 H$. (b) Precession frequency and (c) inverse lifetime obtained by fitting plotted as a function of magnetic field. Solid curves are the results calculated using theoretical formula. (d) Precession amplitude plotted as a function of precession frequency. Horizontal line is a guide to eye.

qualitatively explained by the fact that the optical spin torque was induced by interface effects.

3.3 Evaluation of optical spin torque vectors

To evaluate the optical spin vector quantitatively for different thicknesses, the phase of the magnetization precession was analyzed as shown in Figure 4. Figure 4(a) shows the typical time-resolved MOKE signal when a circularly polarized laser pulse was irradiated on the sample. The data were obtained by taking the differences between signals measured with LCP and RCP lights. All data for the phase analysis are provided in Supplemental Material [39]. The large peak due to specular inverse Faraday effect, as mentioned above, is used to define $t = 0$, the time when pump pulse arrives. Figure 4(b) shows the subsequent magnetization precession. The signal is decomposed into the cosine and sine components by using the following fitting function:

$$f(t) = (a_1 \cos(2\pi ft) + a_2 \sin(2\pi ft)) \exp(-t/\tau), \quad (1)$$

where a_1 and a_2 are the cosine and sine component amplitudes, respectively. They are shown as red and blue solid curves in Figure 4(b), respectively. If the magnetization precession is assumed to be induced by the out-of-plane spin-transfer torque ($\mathbf{m} \times (\mathbf{m} \times \mathbf{s})$), the magnetization is simultaneously tilted away from the film plane by the femtosecond laser pulse and the precession starts from the magnetization tilted away from the film plane, which leads to a cosine signal in the out-of-plane component of magnetization. On the other hand, if magnetization precession is excited by the field-like torque ($\mathbf{m} \times \mathbf{s}$), magnetization is tilted toward in-plane and precession starts

subsequently, leading to a sine signal. Note that the easy axis of the magnetization is in the film plane, and the magnetization is parallel to the magnetic field direction in equilibrium. A small in-plane component of the light wave vector is parallel to the magnetization and does not induce torque on the magnetization. The magnetization dynamics excited by the optical spin torque can be described by the Landau–Lifshitz–Gilbert equation as follows:

$$\frac{d\mathbf{m}}{dt} = -\gamma\mu_0 \mathbf{m} \times \mathbf{H}_{\text{eff}} + \alpha \mathbf{m} \times \frac{d\mathbf{m}}{dt} - \tau_{\parallel}(t) \mathbf{m} \times \mathbf{z} - \tau_{\perp}(t) \mathbf{m} \times (\mathbf{m} \times \mathbf{z}), \quad (2)$$

where, the first and second terms are precession torque and damping torque. $\mathbf{z}$ is a unit vector normal to film plane. $\tau_{\perp}$ and $\tau_{\parallel}$ are the out-of-plane and in-plane optical spin torques, respectively. $\mathbf{H}_{\text{eff}}$ is a effective magnetic field due to external magnetic field and anisotropy field, which is given by,

$$\mathbf{H}_{\text{eff}} = \mathbf{H} - M_{\text{eff}} (\mathbf{m} \cdot \mathbf{z}) \mathbf{z}. \quad (3)$$

By solving the linearized Landau–Lifshitz–Gilbert equation, the following equation is obtained for the out-of-plane component of the magnetization:

$$m_z(t) = \left(\tau_{\perp} \cos(2\pi ft) + \sqrt{\frac{H}{H + M_{\text{eff}}}} \tau_{\parallel} \sin(2\pi ft) \right) \Delta t \times \exp(-t/\tau), \quad (4)$$

where m_z is the out-of-plane component of the normalized magnetization. Here, the phase delay due to the damping term was neglected since α is small enough ($\tan^{-1}(\alpha) = 0.6$ deg. when $\alpha = 0.01$ is used). Δt is the pulse duration, and it is considerably shorter than the period of the magnetization precession, i.e., $2\pi f \Delta t \ll 1$. Therefore, the time-integrated out-of-plane torque $\tau_{\perp} \Delta t$ and in-plane torque $\tau_{\parallel} \Delta t$ can be evaluated by using Eqs. (1) and (4). It should be noted that the effect of $\tau_{\parallel}$ on m_z is increased with increasing H because the elliptical shape of magnetization precession is suppressed by the bias magnetic field. Figure 4(c) and (d) show the two-dimensional plots of the optical spin torques, $\tau_{\perp} \Delta t$ and $\tau_{\parallel} \Delta t$. It was found that the torque vectors depend on both d_{Pt} and d_{FeCo} . The d_{Pt} and d_{FeCo} dependences of the out-of-plane torque and in-plane torque are summarized in Figure 4(e) and (f).

3.4 Discussions of the thickness dependence of the optical spin torques

To discuss the optical spin torque with different d_{Pt} and d_{FeCo} , light absorption and Poynting vectors are calculated

Figure 3: (a) Pt and (b) FeCo thickness dependence of optical helicity-induced magnetization precession for FeCo/Pt bilayer. The sample ($d_{\text{Pt}}, d_{\text{FeCo}} = (2, 3)$ nm) was measured twice.

Figure 4: (a) MOKE signal as a function of pump-probe delay time when circularly polarized laser pulse was irradiated on FeCo/Pt thin films. The optical helicity-induced signal was observed at zero-delay time. (b) Subsequent optical helicity-induced magnetization precession. Dashed curves are fitted results and decomposed into cosine and sine components, shown as red and blue solid curves, respectively. (c) Pt and (d) FeCo thickness dependence of two-dimensional optical spin torque in FeCo/Pt thin films extracted from the phase analysis. (e) Pt and (f) FeCo thickness dependence of the out-of-plane [square symbols] and in-plane torque [triangle symbols]. Dashed curves are calculated normalized light absorption in Pt layer. Solid line in (f) represents in-plane torque proportional to $1/d_{FeCo}$. MOKE, magneto-optical Kerr effect.

with the refractive index values taken from the literature [40–43]. The details of the calculation are provided in the Supplemental Material [39] (see Supplemental Material, Sections I and II). The out-of-plane and in-plane torques are discussed based on the optically generated spin and spin-transport in bilayer thin films, analogous to electrical generation of spin-orbit torque [4, 5, 44–52]. The d_{Pt} and d_{FeCo} dependences of out-of-plane torque can be understood by the fact that spin generated by the optical orientation in Pt layer induces out-of-plane spin-transfer torque. The d_{Pt} dependence of out-of-plane torque shown in Figure 4(e) is similar to light absorption in Pt layer [dashed curve in Figure 4(e)], indicating that spin is generated by light absorption in Pt layer. In addition, the out-of-plane torque increased with decreasing d_{FeCo} , as indicated in Figure 4(f). This is consistent with the nature of the interface effect of the spin-transfer torque. The spin-angular momentum conversion efficiency in Pt layer calculated by

using the light absorption in Pt layer was found to be approximately 2% (see Supplemental Material, Section V [39]). This value is roughly consistent with that reported previously [19].

The in-plane torque can be potentially induced by the following three mechanisms: spin-rotation at the interface, inverse Faraday effect in the ferromagnetic layer, and spin generation via interface spin-orbit coupling. The spin is slightly rotated when it travels across the nonmagnet/ferromagnet interface, which can be described by the imaginary part of the spin-mixing conductance at the interface [4, 5]. However, if the in-plane torque is assumed to be due to the imaginary part of the spin-mixing conductance, the d_{Pt} dependence of in-plane torque should exhibit the same behavior as the out-of-plane torque. In addition, the imaginary part of the spin-mixing conductance is smaller than the real part by a factor greater than ten [53]. These facts rule out the possibility of spin-rotation at the interface. The second

possible consideration is that the in-plane field-like torque is induced by the magnetic field generated by the inverse Faraday effect. In the inverse Faraday effect, the magnetic field generated by the circularly polarized light is proportional to the square of the electric field amplitude $\mathbf{E} \times \mathbf{E}^*$. However, the significant increase of the in-plane torque with decreasing FeCo thickness cannot be explained by the inverse Faraday effect inside the FeCo layer (see Supplemental Material, Section IV). Hence, the above fact cannot explain the d_{FeCo} dependence of in-plane torque [Figure 4(f)] and rules out the second scenario.

The remaining possible mechanism is that the in-plane torque is induced by the spin generated due to the interface spin–orbit coupling because the in-plane field-like torque is inversely proportional to d_{FeCo} [Solid line in Figure 4(f)]. If the stacking structure exhibits inversion symmetry breaking, spin is generated due to Rashba spin–orbit coupling. Optically induced spin via Rashba spin–orbit coupling has been theoretically discussed in previous studies [30, 54–57]. Here, we consider the Rashba spin–orbit coupling with inversion symmetry breaking to be normal to the film plane,

$$\mathcal{H}_R = \frac{\alpha_R}{\hbar} (\boldsymbol{\sigma} \times \mathbf{p}) \cdot \mathbf{z}, \quad (5)$$

where α_R , $\boldsymbol{\sigma}$, and $\mathbf{p}$ are the Rashba parameter, Pauli spin matrix, and electron momentum, respectively. Edelstein derived the optically induced spin due to Rashba spin–orbit coupling as [30],

$$\mathbf{s}_R = iK\mathbf{z}(\mathbf{z} \cdot \mathbf{E} \times \mathbf{E}^*), \quad (6)$$

where we used K as a coefficient of the induced spin $\mathbf{s}_R$. Note that the spin is generated parallel to the film normal, and its sign does not depend on the polarity of the symmetry. The exchange coupling between the magnetization and the spin generated via Rashba spin–orbit coupling induces the in-plane torque $\mathbf{m} \times \mathbf{s}_R$ [51, 52], which is analogous to the electrical manipulation of magnetization via the Rashba–Edelstein effect.

To validate the optical Rashba–Edelstein effect, the optical helicity-induced magnetization precession with a changing stacking structure symmetry (Pt/Co/Pt, Pt/Co/Ta, and Co/Pt) was measured [39] (see Supplemental Material, Section VIII). The in-plane torque was enhanced when Co was sandwiched by SiO_2 and Pt. This confirms that the in-plane torque is induced by the interface Rashba effect due to the asymmetric heterostructure. In addition, the electrical interfacial field-like spin–orbit torque due to the Rashba–Edelstein effect has been reported in similar stacking structures (oxide/[Co, CoFe, CoFeB]/Pt) [47, 49, 58]. This supports the presence of Rashba spin–orbit coupling in the heterostructures used in this study.

Finally, the in-plane torque $\tau_{\parallel}\Delta t$ obtained from the experiments are discussed quantitatively in terms of optical Rashba–Edelstein effect. The in-plane torque in the unit of the angular momentum per unit area $M_s d_{\text{FeCo}} \tau_{\parallel} \Delta t / \gamma$, which was obtained by the slope of $\tau_{\parallel} \Delta t$ vs $1/d_{\text{FeCo}}$ in Figure 4(f), was evaluated to be $\sim 4 \times 10^{-18} [\text{J} \cdot \text{s} \cdot \text{m}^{-2}]$. This value agrees with the calculated value $\sim 2 \times 10^{-18} [\text{J} \cdot \text{s} \cdot \text{m}^{-2}]$ [39] (see Supplemental Material, Section VII), in which the Rashba parameter and the exchange coupling constant are taken from literature [44, 59, 60].

4 Conclusion

This study investigated the optical spin torque vector in FeCo/Pt heterostructure with different thicknesses. d_{Pt} and d_{FeCo} dependences of the out-of-plane and in-plane optical spin torque were evaluated by analyzing the magnetization precession phase. It was shown that the in-plane field-like optical spin torque significantly increased with decreasing d_{FeCo} , indicating the interfacial nature of the torque. The in-plane field-like torque was induced by the spin generated due to the interface spin–orbit coupling as a result of the optical Rashba–Edelstein effect. The optical generation of spin via optical Rashba–Edelstein effect represents a new method of manipulating thin film nanomagnets. However, the field-like optical spin torque induced by the interfacial spin is small compared with the spin-transfer torque induced by the optical orientation of Pt. The materials that are known to show Rashba interface, such as Ag/Bi interface [59, 61, 62], may prove valuable in increasing the efficiency of the interface optical spin generation.

Author contribution: S.I. and S.M. conceived the research idea. S.I. performed the sample fabrication. S.I. and K.I. performed the optical measurement. S.I. performed the data analysis. S.I. and S.M. performed theoretical calculations. All authors contributed to discussions of the results.

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