

Enhancing optically induced intersite spin transfer in antiferromagnetic-ferromagnetic ultrathin films

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(Received 6 August 2024; accepted 23 June 2026; published 27 July 2026)

Antiferromagnetic (AFM) films play a crucial role in advanced spintronic devices by regulating adjacent ferromagnetic (FM) layers via interfacial exchange coupling. However, the impact of antiferromagnets on magnetic dynamics of FM films under ultrashort laser pulses remains largely unknown. Using ultrafast extreme ultraviolet high-harmonic pulses for element-specific magnetization probing, we investigate the femtosecond laser-induced magnetic dynamics in an ultrathin FM Co film with paramagnetic (PM) Cu capping versus AFM Mn capping. Surprisingly, we observe notably shorter demagnetization time for Co in the AFM Mn/FM Co system, approaching the excitation laser pulse duration. During the femtosecond demagnetization process of Co, Mn magnetization arises immediately, indicating a dominant photon-driven interfacial spin-transfer process consistent with optically induced intersite spin transfer (OISTR). Experimental results, supported by time-dependent density functional theory, reveal an increased OISTR magnitude at the Mn/Co interface by a factor of about 1.8 compared to the Cu/Co interface. Furthermore, the photon-driven interfacial contribution saturates at ~ 1 nm, consistent with the short-range nature of OISTR. Remarkably, by utilizing AFM Mn as the adjacent material and adjusting its thickness, OISTR can contribute to $\sim 45\%$ of the demagnetization in ultrathin FM films. The combination of OISTR with ultrathin materials paves the way for next-generation petahertz optospintronics.

DOI: [10.1103/zvd1-qm8s](https://doi.org/10.1103/zvd1-qm8s)

I. INTRODUCTION

Manipulating spin states in magnetic materials within an extremely short timescale is essential for advancing the next generation of spintronic devices [1]. Hence, gaining a comprehensive understanding of the fundamental physical mechanisms governing the dynamics of magnetic materials, with potential applications on ultrafast timescales, is crucial. Among magnetic materials, antiferromagnets possess rich physical properties and promising application prospects. Despite the absence of macroscopic magnetization, the strong exchange coupling between the antiferromagnetic (AFM) sublattices enhances the effective field and characteristic frequency, imparting speed and robustness to antiferromagnets

under external magnetic fields [2,3]. When excited by an ultrafast laser, the AFM-ferromagnetic (FM) coupling exhibits decoupling behavior on the subpicosecond scale [4]. Additionally, the AFM layer can reduce the laser-induced demagnetization time of the adjacent FM layer [5] by allowing a greater penetration depth of the superdiffusive spin current [6,7] into the AFM-ordered layer. Moreover, transient magnetic signals could also occur in antiferromagnets under laser excitation [8,9]. These unique properties make antiferromagnets a cornerstone for the development of ultrafast spintronics [10,11]. However, to date, the speed limit of controlling magnetic dynamics on or via antiferromagnets under the excitation of ultrashort laser pulses remains unclear.

Several spin-transfer mechanisms have been proposed to contribute to ultrafast magnetic dynamics on femtosecond timescales. However, the microscopic origin of ultrafast demagnetization remains an active topic of investigation because multiple spin-transfer and relaxation processes can occur on comparable femtosecond timescales. Besides incoherent channels such as Elliott-Yafet spin-flip scattering [12] and superdiffusive spin transport [5–7,13–16], laser excitation can also generate ultrafast photon-driven spin transfer. In particular, ballistic transport may occur before substantial

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scattering takes place and can therefore contribute to the earliest stages of demagnetization [17,18]. More recently, optically induced intersite spin transfer (OISTR) has been proposed as a photon-driven mechanism involving spin-selective charge transfer between neighboring atoms [19]. Although ballistic transport and OISTR may exhibit similarly rapid temporal responses, occurring on timescales comparable to the duration of the exciting pulse, their characteristic spatial scales are expected to differ substantially. Ballistic transport can extend over tens to hundreds of nanometers, whereas OISTR is generally expected to remain localized within a few atomic layers near magnetic interfaces. Although substantial progress has been made in demonstrating OISTR-related behaviors in half-metallic Heusler materials [20,21], NiFe and CoPt alloys [22,23], and Ni/Pt, Co/Cu, and Mn/Co multilayer films [9,24–26], how adjacent film type and thickness influence OISTR in ultrathin magnetic systems has yet to be explored, leaving this aspect still unclear.

In this work, we present a series of key advances in the understanding and manipulation of ultrafast OISTR in ultrathin magnetic films. By combining temporal and thickness-dependent analyses, we quantified the photon-driven short-range interfacial contribution to Co demagnetization, reaching up to 34% in a Cu/10-monolayer (ML) Co/Cu(001) system, consistent with an OISTR-dominated mechanism revealing its essential role in few-atomic-layer magnetic systems. Remarkably, we discovered that substituting the PM Cu capping layer with AFM Mn enhances this photon-driven short-range contribution to demagnetization by an additional 11%, reaching a total of $\sim 45\%$. Consequently, we observed that the laser-induced demagnetization time of Co moments is significantly shorter in AFM Mn/Co films compared to PM Cu/Co films, highlighting the potential to engineer OISTR coefficients (defined here as the fractional photon-driven short-range contribution to total demagnetization) by modifying adjacent layers. We have also identified a short-range interfacial contribution that saturates at ~ 5 atomic layers (~ 1 nm), consistent with the expected spatial scale of an OISTR-dominated mechanism. Furthermore, these experimental findings are well supported by *ab initio* time-dependent density functional theory (TDDFT) calculations, which show a twofold increase in OISTR at the AFM Mn/Co interface compared to the Cu/Co interface, significantly accelerating Co film demagnetization. Our research not only provides quantitative insights into photon-driven spin-transfer processes at magnetic interfaces but also paves the way for interface-engineered optospintronic devices.

II. RESULTS

A. TR-RMOKE experiments

Figure 1 depicts the schematic of the experimental setup of time-resolved resonant magneto-optical Kerr effect (TR-RMOKE) spectroscopy measurements utilized for measuring the magnetic dynamic responses of Co and Mn following laser excitation. (Please refer to Sec. S1 of the Supplemental Material [27] and Refs. [28–34] for sample preparation. Details about the entire experimental system are provided in Fig. S1, while RMOKE reflecting Kerr rotation [35,36] is presented in Sec. S2.) The ultrathin sample was initially

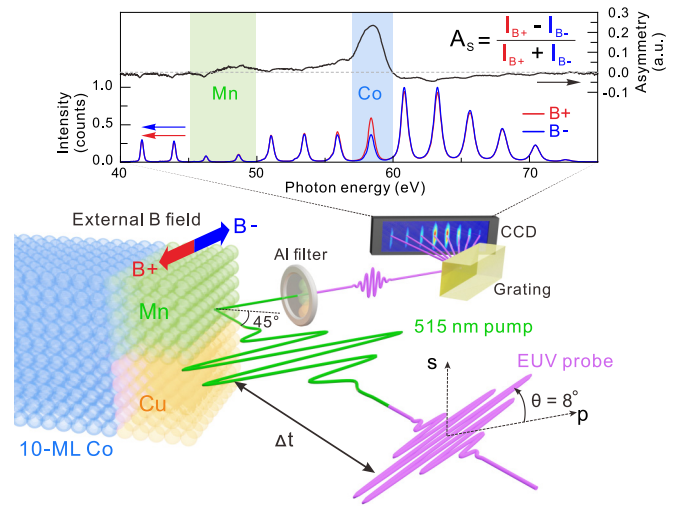


FIG. 1. TR-RMOKE in a longitudinal geometry for probing the magnetic dynamics of Cu/Co vs AFM Mn/Co. The samples were initially excited by a 515 nm pulse laser with a pulse duration of 25 fs in full width at half maximum (FWHM), and subsequently probed by a Δt -delayed linearly polarized EUV pulse with an optimized azimuthal angle 8° relative to the p axis. The top inset shows an example of the high-harmonic generation spectra under the magnetic fields of ± 800 Oe ($B+$ and $B-$) and the corresponding magnetic asymmetry, $A_s = (I_{B+} - I_{B-}) / (I_{B+} + I_{B-})$, of an AFM Mn/Co sample. The green and blue regions mark the M-absorption edges of Mn (45–50 eV) and Co (57–60 eV), respectively.

excited by linearly polarized 25 fs pulses at 515 nm [37]. Subsequently, the variation in the sample magnetization was probed using extreme ultraviolet (EUV) pulses ranging from 40 to 72 eV [38], covering the ranges of M-absorption edges of Mn (45–48 eV) and Co (57–60 eV). Note that an additional 3-ML Cu layer was capped onto the AFM Mn layer to prevent oxidation during long-term measurements. The capping layer is not depicted in Fig. 1.

To distinguish how different capping layers influence the spin dynamics of the FM Co layer, Fig. 2(a) directly compares the energy-integrated magnetization dynamics for Co in the Cu/Co (3-ML Cu/Co) versus Co and Mn in the Mn/Co (3-ML Cu/7-ML Mn/Co) system, all subjected to the same laser excitation with a fluence of 9 mJ/cm^2 . In the Cu/Co system, clear demagnetization of Co was observed upon laser excitation. Remarkably, incorporating the Mn film as the capping layer not only increases the demagnetization level but also reduces the demagnetization time of the Co layer. While Co magnetization is substantially reduced upon light exposure, Mn magnetization arises immediately, indicating the existence of light-induced magnetic moment transfer from Co to Mn, thus unveiling the essential role of OISTR in the demagnetization process of Co. To gain further insights into the spin dynamics in the AFM Mn/Co sample, we also analyze relative variations in the energy-resolved magnetic moment. Figure 2(b) displays the magnetic asymmetry curves extracted 1 eV above and below the Fermi level (E_F) of Co (58 eV) and Mn (47.2 eV) [39]. In Co, immediately upon excitation by light, both the magnetic asymmetry curves above and below the E_F decrease. In contrast, in Mn, during the laser pulse, the

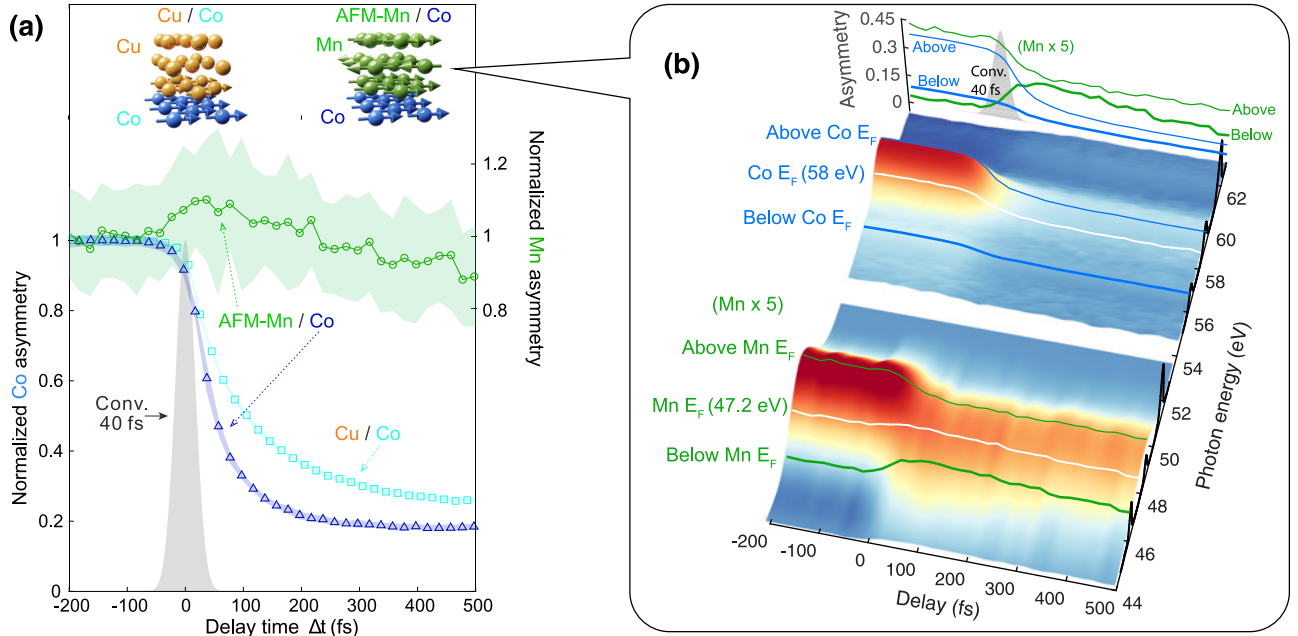


FIG. 2. Experimental magnetization dynamics. (a) Energy-integrated magnetization dynamics for Mn and Co in the Mn/Co vs Co in Cu/Co after excitation of a pulse with a pulse duration of 25 fs and a fluence of 9 mJ/cm². The standard deviation (shaded areas) shown here is based on 20 experimental trials. The dark gray area indicates the convolution, 40 fs in FWHM, of the laser pulse duration (25 fs) and the EUV pulse duration (≈30 fs). (b) The time-dependent magnetic asymmetry spectra of the Mn/Co. The magnetic asymmetry curves extracted 1 eV above (thin lines) and 1 eV below (thick lines) the E_F (white lines) of Co (58 eV) and Mn (47.2 eV) are displayed, as shown in the top inset for comparison.

magnetic asymmetry curve above the E_F transiently decreases, while that below the E_F immediately increases, as summarized in Table I. Such element- and energy-dependent magnetic asymmetry curves are associated with the excited electrons that produce spin current through the Mn/Co interface, combined with spin-flip transitions of the remaining localized electrons, which will be discussed later.

B. OISTR contribution in ultrafast magnetism

A three-exponential phenomenological model, including one demagnetization and two remagnetization terms (one fast

and one slow), was recently proposed for describing ultrafast laser-induced magnetization dynamics of bulk samples [40]. However, it fails to depict the demagnetization curves of ultrathin magnetic multilayer films studied in the present work, due to the significant differences between the previous and present demagnetization processes. Thus, to quantify the OISTR contribution in ultrafast magnetism induced by light in the present systems, we extend this phenomenological model [40] by adding an extra demagnetization term. This results in a four-exponential model, given by

$$M(t_d)/M_0 = G(t_d) \otimes \left\{ 1 + [A_{\text{demag},1} e^{-\frac{-(t_d-t_0)}{\tau_{\text{demag},1}}} + A_{\text{demag},2} e^{-\frac{-(t_d-t_0)}{\tau_{\text{demag},2}}} - A_{\text{remag},1} e^{-\frac{-(t_d-t_0)}{\tau_{\text{remag},1}}} - A_{\text{remag},2} e^{-\frac{-(t_d-t_0)}{\tau_{\text{remag},2}}}] \theta(t_d - t_0) \right\}, \quad (1)$$

where $M(t_d)$ is the magnetization varying with delay time t_d . M_0 corresponds to the magnetization before laser excitation. The Gaussian profile $G(t_d)$ represents the temporal resolution of our RMOKE system. $A_{\text{demag},1}/\tau_{\text{demag},1}$ and $A_{\text{demag},2}/\tau_{\text{demag},2}$ represent the amplitude and time constant of the two demagnetization channels, respectively; $A_{\text{remag},1}/\tau_{\text{remag},1}$ and $A_{\text{remag},2}/\tau_{\text{remag},2}$ correspond to the amplitude and time constant of the two remagnetization channels, respectively. The relationship $A_{\text{demag},1} + A_{\text{demag},2} = A_{\text{remag},1} + A_{\text{remag},2}$ holds because the magnetization fully recovers at long times. $\theta(t_d - t_0)$ is Heaviside step function, starting at the delay time t_0 . Figure 3(a) shows the experimental data of the laser-induced magnetic dynamics of Co in Cu/Co and

TABLE I. Transient magnetic moment (ΔM) in the Mn/Co/Cu induced by the excitation laser.

		Cu ^a	Co	Mn
ΔM in Exp. Fig. 2	total		↓ ^b	↑ ^c
	above E_F		↓	↓
	below E_F		↓	↑
ΔM in TDDFT Cal. (noSOC)	total	↑	↓	↑
	above E_F	↑	↓	↓
	below E_F	↑	↑	↑
ΔM in TDDFT Cal. (SOC)	total	↑	↓	↑
	above E_F	↑	↓	↓
	below E_F	↑	↓	↑

^aNo RMOKE signal for Cu in this experiment.

^bDecrease.

^cIncrease.

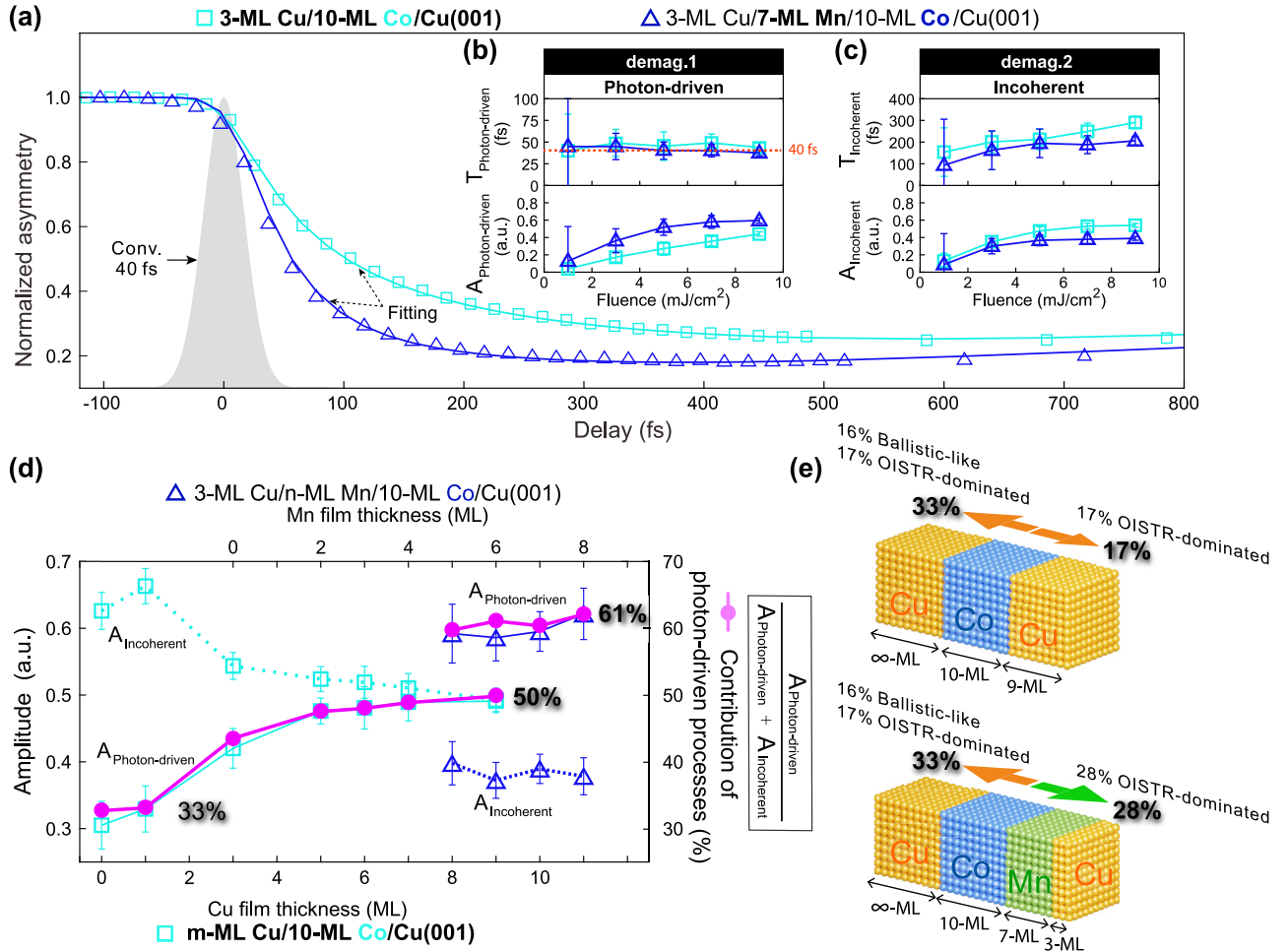


FIG. 3. Quantifying photon-driven spin-transfer contributions to Co demagnetization. (a) Experimental observations (cyan squares for Mn/Co and blue triangles for Cu/Co) and their corresponding fitting curves (solid lines) for Co demagnetization after excitation with a 25 fs, 515 nm pulse at a fluence of $9 \text{ mJ}/\text{cm}^2$, showing faster demagnetization for the Mn-capped sample. The fitting includes two demagnetization and two remagnetization terms. The gray-shaded area represents the pulse convolution (~ 40 fs). Insets (b) and (c) display two distinct demagnetization terms under varying fluences, distinguishing between photon-driven and incoherent processes. (b) Extracted amplitudes ($A_{\text{Photon-driven}}$) and time constants ($\tau_{\text{Photon-driven}}$) for the direct photon-driven demagnetization term (demag.1) as a function of fluence, highlighting directly photon-driven process with $\tau_{\text{Photon-driven}} \approx 40$ fs. (c) Amplitudes ($A_{\text{Incoherent}}$) and time constants ($\tau_{\text{Incoherent}}$) for the incoherent demagnetization term (demag.2) as a function of fluence, associated with secondary processes (e.g., spin-flip scattering, superdiffusive spin currents). (d) $A_{\text{Photon-driven}}$ and $A_{\text{Incoherent}}$ (left axis), along with their contribution percentages (right axis), as a function of capping layer thickness (n for Mn and m for Cu layers) at a fixed pump fluence of $9 \text{ mJ}/\text{cm}^2$. The data show saturation of $A_{\text{Photon-driven}}$ or its contribution with increasing Cu capping layer thickness, reaching ~ 5 ML, and a higher contribution of $A_{\text{Photon-driven}}$ from Mn capping (61%) compared to Cu (50%). (e) Schematic illustrating OISTR and ballistic spin transport contributions in Cu and Mn capping systems. Refer to the main text for details.

AFM Mn/Co films as well as the fitted curves derived from Eq. (1). The fitted results show excellent agreement with demagnetization with a delay time of up to a few picoseconds (see Fig. S2, Sec. S4, and Fig. S10 in the Supplemental Material [27]).

Figure 3(b) presents the amplitudes and time constants extracted from the fitted demagnetization term (demag.1) as a function of pumping fluence. Notably, across all independent experiments conducted with varying pumping fluences, the fitted time constants ($\tau_{\text{demag.1}}$) cluster around 40 fs, closely matching the convolution duration of the laser pulse duration (25 fs) and the EUV pulse duration (≈ 30 fs). This observed behavior strongly suggests that the underlying mechanism of demag.1 term is fully optically driven and governed by

the characteristics of the excitation pulses. We thus attribute this term to a direct photon-driven process, involving directly photon-excited spin-selective electrons, with its time constant and amplitude denoted as $\tau_{\text{Photon-driven}}$ and $A_{\text{Photon-driven}}$, respectively. According to Ref. [19], these features are consistent with the characteristics of the OISTR mechanism. Furthermore, $A_{\text{Photon-driven}}$ exhibits a linear increase with laser fluence and reaches saturation at higher fluence levels, which could be attributed to the finite number of available states above E_F . Notably, $A_{\text{Photon-driven}}$ may also include contributions from ballistic spin transport [39,40], which occurs without scattering events (unlike superdiffusive spin currents, a secondary process involving multiple scattering events). However, these two direct photon-driven processes, OISTR and ballistic spin

transport, are expected to exhibit fundamentally different spatial characteristics. This distinction will be further analyzed through the results of the thickness-dependent experiments, with the discussion starting in the paragraph after the next two paragraphs.

On the other hand, Fig. 3(c) displays another set of amplitudes and time constants (demag.2). In contrast to $\tau_{\text{demag.1}}$, $\tau_{\text{demag.2}}$ ranges from ~ 100 to ~ 300 fs as the pumping fluence increases. According to the literature [41], the fluence-dependent magnetic responses at this timescale are associated with incoherent secondary processes, such as Elliott-Yafet electron-phonon scattering [12], electron-magnon scattering [42], Coulomb exchange scattering [43], and superdiffusive spin current [5–7,13–16]. The overlapping timescales of these mechanisms make definitive attribution challenging. Therefore, we designate this slower demagnetization term as arising from incoherent secondary processes, collectively represented by $\tau_{\text{Incoherent}}$ and $A_{\text{Incoherent}}$. A clear trend of increasing $\tau_{\text{Incoherent}}$ with rising pump fluence is observed, aligning with previous reports [12,44] that attribute this behavior to enhanced spin fluctuations at elevated spin temperatures induced by higher fluences [45]. Correspondingly, $A_{\text{Incoherent}}$ shows a saturation phenomenon at high fluences, indicating limited demagnetization channels available at this time constant.

At a fixed pump fluence of 9 mJ/cm^2 , we conducted a series of experiments and analyses incorporating different capping elements (Cu and Mn) with varying thicknesses to compare the contributions of direct photon-driven processes and incoherent secondary processes from these films, as depicted in Fig. 3(d). This fluence level, 9 mJ/cm^2 , is sufficiently high to completely quench the magnetization of Co, i.e., $A_{\text{demag.1}} + A_{\text{demag.2}} \approx 1$ (or $A_{\text{Photon-driven}} + A_{\text{Incoherent}} \approx 1$). We also computed the ratio of the direct photon-driven demagnetization amplitude to the total demagnetization amplitude, $A_{\text{Photon-driven}} / (A_{\text{Photon-driven}} + A_{\text{Incoherent}})$, to quantify the role of the direct photon-driven process in laser-induced demagnetization. It is also important to note that capping layers with only a few atomic thicknesses applied in these experiments have a negligible effect on the 515 nm pump absorption in the 10-ML Co layer (see Sec. S3 of the Supplemental Material [27] for layerwise absorption analysis using the transfer matrix method [46–49]). This confirms that the variation in demagnetization amplitude with capping thickness, as depicted in Fig. 3(d), arises from OISTR effects at the capping layer-Co interface rather than changes in Co absorption induced by the capping layer.

Next, we analyze the exact contribution of OISTR to demagnetization. In the absence of a capping layer [bare 10-ML Co/Cu(001)], the direct photon-driven process contributes to demagnetization with a timescale of 40 fs and accounts for 33% of the total demagnetization. The remaining 67% occurs on a longer timescale of 150–300 fs through incoherent secondary processes. When a Cu capping layer is added on top of the Co/Cu(001), the contribution of the photon-driven process to demagnetization noticeably increases, while that of the incoherent process decreases correspondingly. This enhancement can result from either ballistic spin transport or OISTR. Although OISTR and ballistic spin transport can both contribute on pulse-duration-limited timescales, their differ-

ent spatial characteristics—ballistic transport spans distances beyond 100 nm [17,18] while OISTR is confined to much shorter ranges [19]—allow their dominant contributions to be separated through thickness-dependent measurements. With sufficient pump photon density as provided in our experiment, ballistic transport would be expected to continuously increase the direct photon-driven demagnetization term as more final states become accessible. However, Fig. 3(d) reveals that this increase saturates at ~ 5 Cu layers, which is inconsistent with the ballistic transport behavior. Thus, the saturation identifies a short-range contribution consistent with an OISTR-dominated interfacial channel. Figure 3(d) also shows that beyond 5 Cu layers, the signal continues to increase but at a much slower rate. Specifically, the first ~ 5 Cu layers at the Cu/Co interface contribute $\sim 17\%$ short-range OISTR-dominated contribution to the instantaneous demagnetization of Co. Increasing the Cu thickness further, up to the ~ 2 nm Cu substrate, adds an additional $\sim 16\%$ longer-range ballisticlike contribution. Thus, in a 9-ML Cu/Co/Cu(001) sample, the combined short-range OISTR contributions from the two Cu/Co interfaces ($\sim 17\% + \sim 17\%$) and the long-range ballistic spin transport to the Cu(001) substrate ($\sim 16\%$) account for $\sim 50\%$ of the total direct photon-driven contributions to Co demagnetization [see Fig. 3(e)]. Thus, our findings suggest that OISTR and ballistic spin transport coexist in the direct photon-driven demagnetization process. OISTR dominates within a few atomic layers near the interface with the Co layer, while ballistic spin transport provides long-range spin transfer between the Co film and the thick Cu(001) substrate.

Regarding why OISTR extends beyond a single atomic interface, we speculate that this is due to the hybridization of electronic states. The overlap of electronic wavefunctions facilitates this process, allowing OISTR to influence several atomic layers, with its effect diminishing as the wavefunction overlap decreases [50,51]. In particular, for 3d transition metals, the effective spin correlation length, determined by the scale of spin-dependent wavefunction overlapping, typically ranges from ~ 2.5 to 16 atomic layers [52–54]. This property of electronic hybridization provides a plausible explanation for the spatial influence of the OISTR effect extending to ~ 1 nm.

Surprisingly, when replacing the Cu capping with AFM Mn (3-ML Cu/7-ML Mn; see Sec. S1 and Fig. S4(b) in the Supplemental Material [27] for details on AFM Mn), the overall photon-driven contribution to the Co demagnetization increases to 61% (while incoherent secondary contribution decreases to 39%). This implies that compared to Cu, the AFM-Mn capping layer can further increase 11% in OISTR ($\tau_{\text{Photon-driven}} \approx 40$ fs) and simultaneously decrease 11% in incoherent part ($\tau_{\text{Incoherent}} \approx 250$ fs). This explains why AFM Mn can accelerate the demagnetization of Co, bringing its demagnetization time constant closer to the pulse duration of the pump pulse. It is important to highlight that we have estimated the spatial extent of OISTR to be ~ 5 ML (~ 1 nm) in Cu/Co/Cu(001) system. Moreover, in the Mn/Co/Cu(001) films, the contribution of demag.1 term remains nearly unchanged when the thickness of Mn layer increases from five to eight. This indicates that OISTR at the Mn/Co interface also

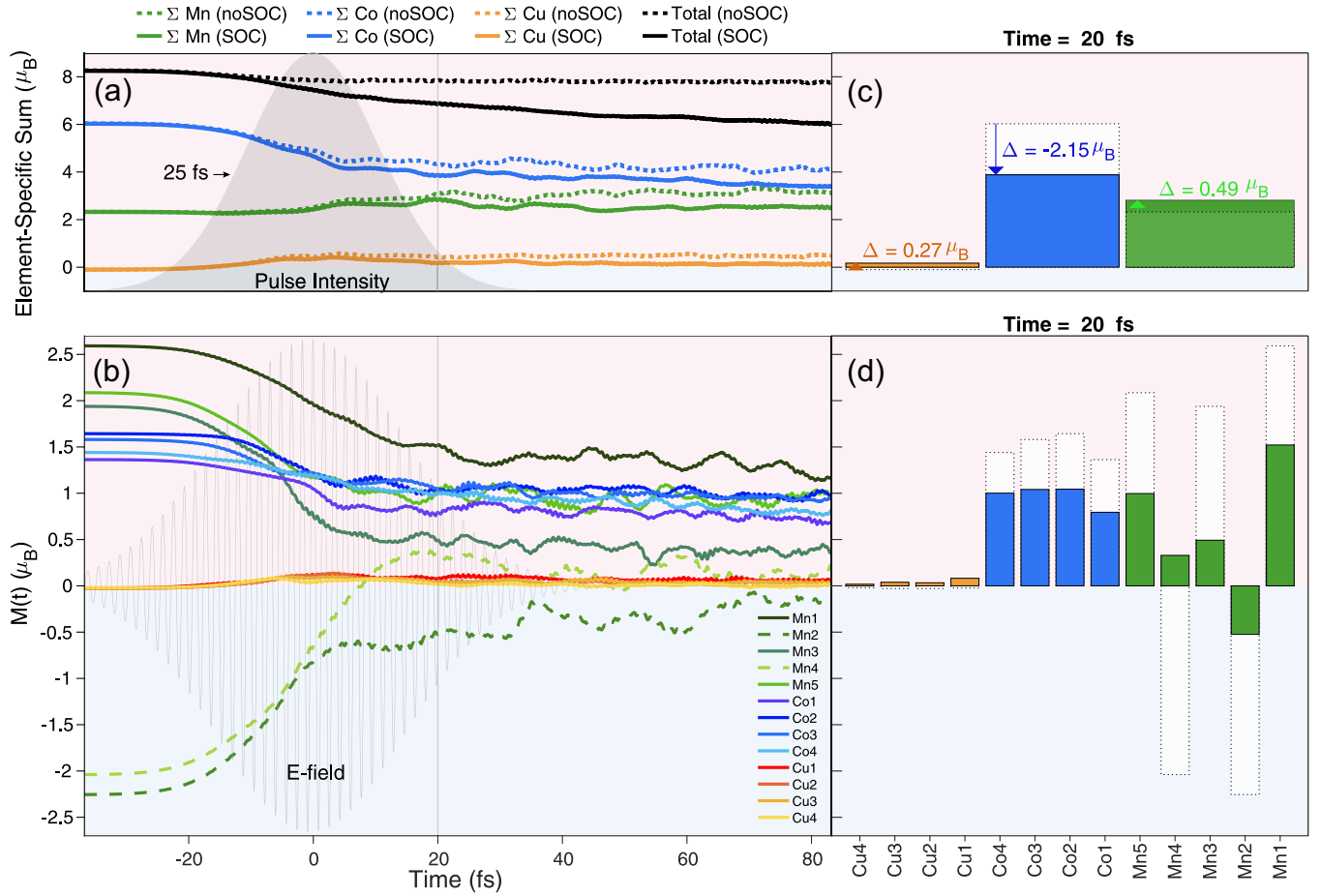


FIG. 4. Calculated time-resolved magnetic moments. TDDFT calculations for (a) element-resolved and (b) layer-resolved magnetic moments of a 5-ML Mn/4-ML Co/4-ML Cu system under excitation by a linearly polarized 515 nm laser pulse with a pulse duration of 25 fs and a fluence of 9 mJ/cm². Snapshots of magnetic moments at a delay of 20 fs are provided for (c) individual elements and (d) each layer. All calculations include the SOC, except for those depicted by dotted lines in panel (a) for comparison.

reaches saturation within this thickness range, consistent with observations in the Cu/Co/Cu(001) system.

C. Spin dynamics by TDDFT calculations

To further elucidate why the AFM Mn films, compared to the PM Cu films, can induce faster demagnetization in the Co layer, we theoretically study the laser-induced ultrafast demagnetization dynamics in the considered Mn/Co/Cu sample by performing the *ab initio* calculations using TDDFT [55,56] with the adiabatic local spin-density approximation [57]. Here, the current TDDFT calculations focus exclusively on the sub-100 fs timescale, where ultrafast electronic dynamics dominate, but do not include incoherent processes such as phonon-mediated Elliott-Yafet spin-flip scattering or superdiffusive spin transport [19], which generally take place on longer timescales (beyond 100 fs or even in the ps regime). In the present calculations, the state-of-the-art all-electron full-potential linearized augmented plane wave method implemented in the ELK code [58] was used. The multilayer structure consists of 5-ML Mn/4-ML Co/4-ML Cu. This simpler structure, composed of a total of 13 atomic layers, adequately describes our experimental sample. The initial spin configuration is determined based on the experi-

mental observations of RMOKE and static density functional theory calculations (see Fig. S7 and Sec. S5 in the Supplemental Material [27]). The Co atoms in the Co slab are ferromagnetically coupled and the Mn atoms in the Mn slab are antiferromagnetically coupled, while the interface between Mn and Co exhibits FM coupling (i.e., Mn and Co spins are parallel). The system is excited using a linearly polarized laser pulse with a wavelength of 515 nm (photon energy of 2.4 eV), a fluence of 9 mJ/cm², and a pulse duration of 25 fs.

Figure 4 illustrates the laser-induced magnetic dynamics of the 5-ML Mn/4-ML Co/4-ML Cu film. The element- and layer-resolved magnetic moments as a function of time are shown in Figs. 4(a) and 4(b), respectively, while Figs. 4(c) and 4(d) highlight their snapshots after the laser excitation. A direct comparison of the experimental observations [Fig. 3(e)] can be made with the *ab initio* calculations [Fig. 4(c)], where after laser excitation, the decrease in the Co magnetization is accompanied by an increase in magnetization in adjacent Cu and Mn layers. This supports the presence of OISTR among them. While the Co magnetic moment decreases by $2.15\mu_B$, the Mn and Cu moments increase by $0.49\mu_B$ and $0.27\mu_B$, respectively. Therefore, the adjacent Mn layer can provide nearly 1.8 times more OISTR compared to the Cu

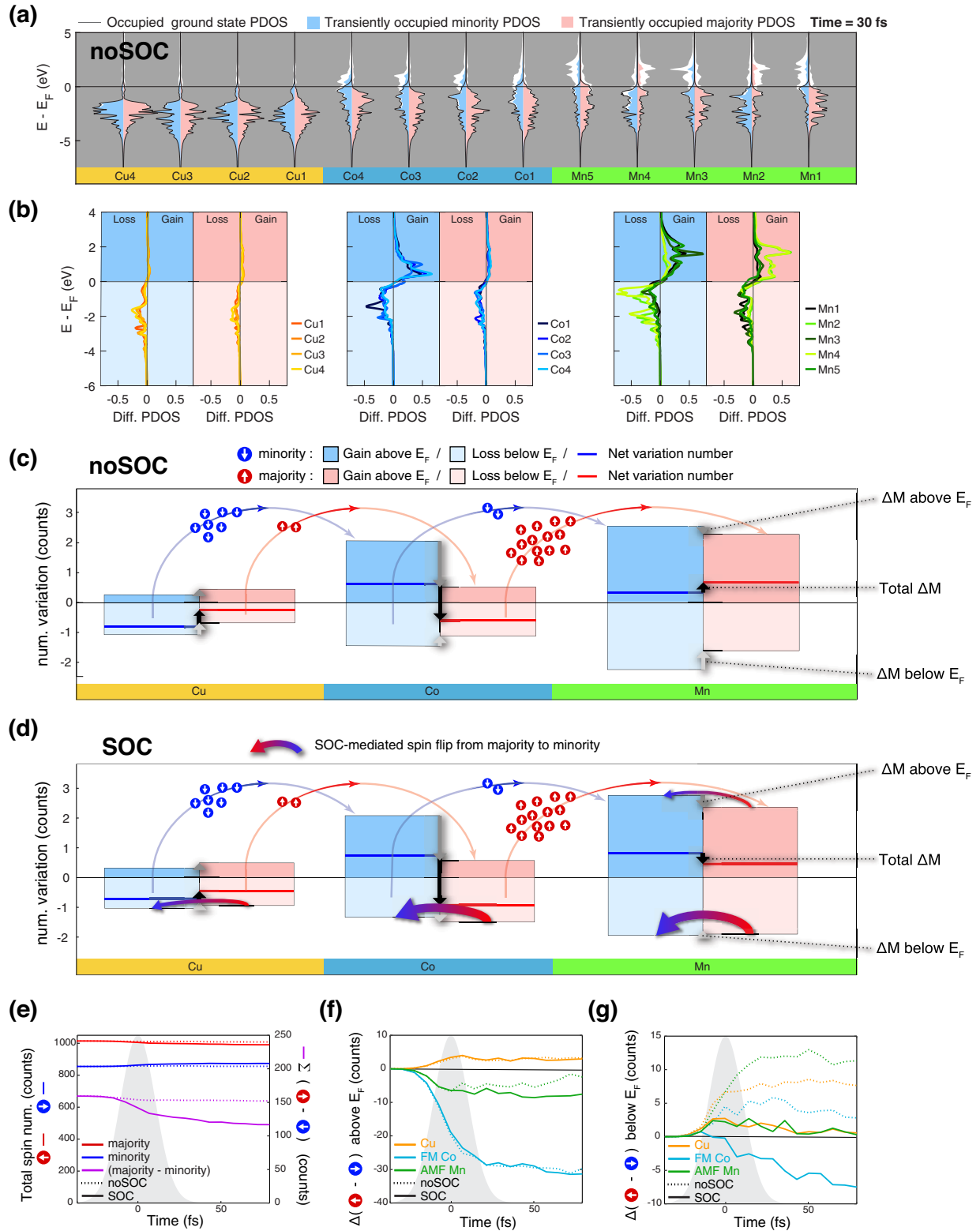


FIG. 5. Laser-induced spin transport in the Mn/Co/Cu system. (a) PDOS of all atomic layers in a 5-ML Mn/4-ML Co/4-ML Cu system, presented in states/eV/spin, at a time delay of 30 fs. (b) Difference in the occupied PDOS between 30 and 0 fs (i.e., the occupied ground state PDOS). (c) Histogram analysis of panel (b) reveals the element- and spin-resolved variations of the electron number below and above E_F , while the thick horizontal bar indicates the net variation number. Additionally, the macroscopic magnetic moment change, ΔM , is associated with the difference in numbers between the spin-up and spin-down electrons. ΔM above and below E_F , as well as the total ΔM , are proportional to the sizes of the dark gray, light gray, and black arrows, respectively. (d) The same as in panel (c) but the SOC included, thus allowing spin flips between the spin-up and spin-down states, as shown in panel (e). SOC-mediated spin flips are weak and only observed in the Mn atoms above E_F , as shown in panel (f), but become prominent in all the atoms below E_F , as depicted in panel (g).

layer, as $(0.49\mu_B)/(0.27\mu_B) = 1.8$, which is very close to our experimental observation of $28\%/17\% = 1.64$ shown in Fig. 3(e). Furthermore, Fig. 4(d) clearly demonstrates that the demagnetization of each Mn layer primarily stems from the OISTR occurring between the Mn layers in the AFM Mn slab. A direct visualization of the *ab initio* simulation without and with spin-orbit coupling (SOC) can be found in Movies S1 and S2, respectively, of the Supplemental Material [27].

Figure 5(a) [and Figs. S3(a)–S3(c) of the Supplemental Material [27]] shows the partial density of states (PDOS) for each layer after the laser excitation without the SOC, where spin-up is set as the global majority here. For comparison between Fig. 5(a) (TDDFT) and the element-specific magnetization above and below E_F [experiments shown in Fig. 2(b) and Table I], Fig. 5(b) illustrates the difference in the occupied PDOS before and after the laser excitation. Figure 5(c) further sums up the variation of the spin-up and spin-down electrons above and below the E_F before and after laser excitation, in which because of the nature of laser excited electron events, the positive (negative) histogram quantifies the total increase (decrease) in the electrons above (below) the E_F . The difference in numbers between spin-up and spin-down electrons under the laser excitation causes (or reflects) the transient changes in magnetic moment.

Based on the conservation of the sum of the spin-up and spin-down electrons before and after excitation, as shown in Fig. 5(e), and by comparing the net variation of the total electron number between the atoms of different elements, Fig. 5(c) concludes that at the Cu/Co interface, the spin current mainly comprises spin-down electrons (as minority spin in Co) flowing from Cu to Co. This is because Cu has nearly no unoccupied DOS available above the E_F [see Fig. 5(a)]. As a result, both spin-up and spin-down electrons tend to flow into the Co slab where unoccupied DOS exist above the E_F . Moreover, there are more unoccupied DOS for the spin-down electrons in Co, thus generating a spin-selective current. Interestingly, at the Mn/Co interface, the adjacent AFM Mn film with very high exchange splitting (AFM ordering) in these Mn sublattices, obtaining considerably unoccupied DOS as a spin sink for both spin-up and spin-down electrons. The spin current at the Mn/Co interface primarily comprises spin-up electrons (as majority spin in Co) flowing from Co to Mn, since Co has few unoccupied DOS for the spin-up state above the E_F . Note that for the spin-down (minority) electrons in Co, even though there are still unoccupied DOS above the E_F , some of them still flow into the Mn slab because there are considerably more unoccupied DOS for the spin-down electrons in the AFM Mn [much larger than that Co has, as shown in Fig. 5(a)]. Although the flow of the spin-down electrons here may reduce the magnitude of spin current (the overall magnetic moment transfer) through the AFM Mn/Co interface, both experimental observations and *ab initio* calculations indicate that, compared to the Cu/Co, the Mn/Co has about 1.8 times higher OISTR coefficient.

After including the SOC, Fig. 5(e) illustrates a notable decrease in the total number of the spin-up electrons (while the spin-down electrons increase) due to SOC-mediated spin flips. Figures 5(f) and 5(g) reveal that above the E_F , spin flips occur only minimally within the Mn slab. In contrast, below the E_F , spin flips become rather significant, causing even the expected

increase in the Co magnetic moment to flip and decrease. The transients of overall magnetic moments, and magnetic moments below and above the E_F in both the Mn and Co layers, as summarized in Fig. 5(d) [and Figs. S3(d)–S3(f) of the Supplemental Material [27]], agree well with the experimental observations (Table I). A direct visualization of PDOS after the laser excitation without and with the SOC can be found in Movies S3 and S4, respectively, of the Supplemental Material [27]. The experimental results, corroborated by *ab initio* calculations, for the first time clearly demonstrate the involvement of SOC-mediated spin flips in laser-induced magnetic dynamics, particularly influencing magnetic moment behavior below the E_F as previously suggested [59].

III. CONCLUSIONS

Through the TR-RMOKE experiments, quantitative analysis using the four-exponential phenomenological model, and TDDFT calculations, this study clearly elucidates the significance of OISTR in ultrathin FM films. We have found that using an AFM Mn film as the adjacent layer, compared to a PM Cu film, dramatically accelerates the demagnetization of the neighboring FM Co film because of a higher OISTR coefficient at the AFM/FM interface. Specifically, our TDDFT calculations reveal distinct spin-transfer channels at the PM Cu/Co and AFM Mn/Co interfaces. At the PM Cu/Co interface, there is primarily a minority spin inflow into the Co layer, while at the AFM Mn/Co interface, there is mainly a majority spin outflow from the Co layer. The type and magnitude of the spin current provided by OISTR are determined by the available electronic states around the E_F at the interface. Furthermore, when the adjacent layer gets thicker, more electronic states become available, which can enhance the photon-driven interfacial spin-transfer contribution. However, our experiment reveals that the thickness effect saturates beyond ~ 1 nm, given the limited spatial reach of OISTR, thereby establishing a characteristic interfacial length scale for photon-driven spin transfer in ultrathin magnetic heterostructures. The photon-driven demagnetization process contains two distinguishable components: a short-range OISTR-dominated contribution confined to ~ 1 nm near the interface and a longer-range ballisticlike contribution extending beyond. We also verify the early involvement of spin flips in laser-induced magnetic dynamics. This notably impacts the behavior of magnetic moments below the E_F .

Nevertheless, by engineering ultrathin magnetic heterostructures, photon-driven interfacial spin transfer can make a dominant contribution to magnetic dynamics. For instance, the photon-driven short-range contribution reaches $\sim 45\%$ in Mn/Co/Cu, arising from the combined contributions of the Cu/Co ($\sim 17\%$) and Mn/Co ($\sim 28\%$) interfaces, demonstrating that interface engineering provides an effective route for controlling ultrafast magnetic dynamics. We can therefore expect enhanced coherent OISTR in nanoscale magnetic systems with large surface-to-volume ratios, including ultrathin films, low-dimensional materials, nanowires, and nanodots, where interfacial effects become increasingly dominant and may enable more efficient photon-driven spin manipulation.

Finally, since OISTR is triggered almost instantaneously upon light incidence—on subfemtosecond timescales

[24,26]—the combination of ultrashort pulses with ultrathin magnetic films, designed with high OISTR coefficients [60], offers a route toward next-generation petahertz optospintronics and may facilitate future advances in the low-dimensional materials and spintronics communities.

ACKNOWLEDGMENTS

M.-C.C. acknowledges Taiwan's 2030 Young Scholar Fellowship. We gratefully acknowledge funding support by

National Science and Technology Council Taiwan, under Grants No. 113-2112-M-007-042-MY3 to M.-C.C., Grant No. 114-2112-M-018-007 to B.-Y.W., and Grant No. 114-2123-M-002-009 to G.-Y.G.

DATA AVAILABILITY

Raw experimental data (corresponding to Fig. S2 in the Supplemental Material and used to generate Fig. 3 in this article) are available in Ref. [61]. All other study data are provided in the article and/or the Supplemental Material [27].

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