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Optically-induced ultrafast magnetization switching in ferromagnetic spin valves

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The discovery of spin-transfer torque (STT) enabled the control of the magnetization direction in magnetic devices in nanoseconds using an electrical current. Ultrashort optical pulses have also been used to manipulate the magnetization of ferrimagnets at picosecond timescales by bringing the system out of equilibrium. So far, these methods of magnetization manipulation have

been mostly developed independently within the fields of spintronics and ultrafast magnetism. Here we show an optically-induced ultrafast magnetization reversal

taking

place within less than a picosecond in rare-earth-free archetypical spin valves of [Pt/Co]/Cu/[Co/Pt] commonly used for current-induced STT switching. We find that the magnetization of the free layer can be switched from a parallel to an antiparallel alignment, as in STT, indicating the presence of an unexpected, intense and ultrafast source of opposite angular momentum in our structures. Our findings provide a route for ultrafast magnetization control by bridging concepts from spintronics and ultrafast magnetism.

Spin-transfer torque (STT)^{1,2}, resulting from spin angular momentum transfer, has enabled the electrical manipulation of magnetization in solid-state devices. The discovery of STT paved the way for non-volatile memory applications such as spin-transfer-torque magnetoresistive random access memory (STT-MRAM) and led to the discovery of the spin-torque oscillation³ and the spin-torque diode effect⁴, with potential applications in neuromorphic computing⁵ and energy harvesting⁶.

Current-induced bipolar STT switching (i.e., changing the polarity of the switching) occurs due to spin angular momentum transfer between two ferromagnetic layers mediated by spin-polarized conduction electrons in spin valves⁷ and magnetic tunnel junctions⁸. In those devices, magnetization switching can be observed with current pulses down to a few hundred picoseconds of duration, but the critical switching current and consequently the switching energy increases rapidly with decreasing pulse duration⁹. The reason is that the spin angular momentum transfer required to overcome the energy barrier to reverse the magnetization has to take place while applying the current pulse. Thus, a fast magnetization switching below a few hundred picoseconds with low energy consumption by STT is hard to obtain in an equilibrium

regime for which the magnitude of the magnetization remains the same. As a solution, a temporary heating of the magnetic material (head-assisted magnetic recording: HAMR¹⁰) can be used to reduce the energy barrier. In spin-orbit torque (SOT) switching¹¹ using 6 ps electrical pulses generated via photoconductive switches, ultrafast heating was also estimated to be beneficial¹².

Furthermore, ultra-short optical pulses can also be used to manipulate the magnetization direction without any applied magnetic field, a phenomenon which is called all-optical switching (AOS). However, only a few specific systems containing ferrimagnetic materials experience ultrafast single pulse AOS¹³⁻¹⁷. The underlying mechanism of this single pulse AOS is understood as a result of angular momentum transfer between two antiferromagnetically coupled magnetic sublattices via conduction electrons triggered by laser-induced ultrafast demagnetization^{13,18}. Recently, the reversal of the magnetization of a FM layer via single laser pulses has been demonstrated by combining a GdFeCo layer experiencing ultra-fast demagnetization (or AOS) and a FM layer in multilayer heterostructures: exchange-coupled GdFeCo/[Co/Pt] bilayers¹⁹ and GdFeCo/Cu/[Co/Pt] spin-valve structures without exchange coupling between the two magnetic layers²⁰. In the latter case, the switching of the FM's magnetization is accomplished due to the combination of its ultrafast demagnetization and angular momentum transfer from the Gd sub-lattice in GdFeCo mediated by conduction electrons²⁰⁻²³. Interestingly, it has been demonstrated that these processes take place during the first picoseconds^{22,23}. However, there is no report on ultrafast magnetization switching in ferromagnetic spin-valves yet. Moreover, the current understanding²⁴ of AOS typically requires starting with an antiparallel (AP) alignment of the magnetizations in two sublattices (ie: Gd and FM in a spin valve^{20,25}) while, in contrast, STT allows switching when starting from both a parallel (P) or an AP magnetic configurations⁷.

Here we demonstrate the magnetization reversal of a [Co/Pt] ferromagnetic layer induced by a single femtosecond laser pulse within a common [Co/Pt]/Cu/[Co/Pt] perpendicular spin-valve

structure²⁶ (Fig. 1a). At high fluence, we observe a reversal from the AP to the P configuration of the free [Co/Pt] layer (Fig. 1b), while at low fluence we observe a switching from the P to the AP state (Fig. 1c). The P-to-AP switching is observed even with an 80 nm Cu spacer and takes place within less than a picosecond. From static and dynamic measurements, we conclude that the laser-induced ultrafast spin injection governs the magnetization reversal even against the magnetization of the reference layer (Fig. 1c). This strengthens the parallels between STT and ultrafast magnetism, suggesting that the spintronic playground can be further exploited at ultrafast timescales.

First, we show the single-shot switching in the spin valve. We prepared spin valves consisting of [Co(1)/Pt(1)]₃/Co(1)/Cu(t_{Cu})/[Co(0.6)/Pt(1)]₂ (Fig. 1a) in which we varied the Cu thickness from 5 to 80 nm (see Methods for details). Here we define [Co(1)/Pt(1)]₃/Co(1) and [Co(0.6)/Pt(1)]₂ as the reference and free layers, respectively. Figure 2a shows the magneto-optic Kerr effect (MOKE) signal (θ_{K}) as a function of the magnetic field (H) applied perpendicularly to the film plane for the sample with $t_{\text{Cu}} = 10$ nm. One can confirm four magnetic configurations defined as P+, AP+, AP-, and P- states. Figures 2b and Extended Data Figure 1a show MOKE microscopy images obtained after exciting the sample with a single linearly polarized femtosecond laser pulse of fluences $F = 3.7$ and 5.9 mJ/cm² (7.6 mJ/cm² shown in Extended Data Fig. 1a), for both P+ and AP+ initial magnetic configurations. Here, the sample is irradiated from the free layer side and MOKE images are obtained from the reference layer as shown in Fig. 2b. For a fluence of $F = 3.7$ mJ/cm² and starting from a P+ state, we observe a transition from red to blue, corresponding to a single-shot P-to-AP switching. On the contrary, no reversal is observed when the initial state is AP+. Therefore, a laser pulse always stabilizes an AP state at low fluences, regardless of the initial state. On the other hand, we observe the opposite behavior favorizing an AP-to-P switching above a fluence $F = 5.9$ mJ/cm². Above $F = 7.6$ mJ/cm², a multidomain state appears at the center in addition to the two

types of switching due to the higher energy density and the spatial energy distribution of the Gaussian beam as shown in Fig. S3c. The small difference in the threshold fluences for AP-to-P switching and multidomain formation, as depicted in Extended Data Fig. 3a, results in a decrease in the final contrast, which is not exactly that of a P state (70-80% of P state contrast). This indicates a partial switching (Extended Data Fig. 1c and Supplementary Information S1).

Next, we demonstrate single-pulse switching in a spin valve with $t_{\text{Cu}} = 80$ nm, so that the laser pulse cannot excite the reference layer (the calculated energy absorption is shown in Extended Data Fig. 2b). Figure 2c shows MOKE hysteresis loops for the free and reference layers detected from both sides. Figure 2d shows MOKE images after shining a laser pulse on a P+ (red) and AP+ (blue) state with $F = 6.9$ and 12.0 mJ/cm² (profiles are shown in Extended Data Fig. 1c and d). In this experiment, we shine the laser pulse on the free layer with the angle of incidence of approximately 45° and image via polar MOKE on the same side as shown in Fig. 2d. Even though the reference layer is not irradiated, we can still observe a P-to-AP switching, while AP-to-P switching is not observed whatever the laser fluence. We summarize the obtained threshold fluences (absorbed in the free layer) for P-to-AP switching, AP-to-P switching, and multidomain state formation as a function of the Cu thickness t_{Cu} in Extended Data Fig. 3c. The AP-to-P switching is observed up to $t_{\text{Cu}} = 40$ nm while only a multidomain state of the free layer is observed above this value (see Supplementary Information S1). The threshold absorbed fluence for AP-to-P switching behaves more like the one for the multidomain state and is improved with thinner t_{Cu} (see Supplementary Information S1 and S3). On the other hand, the threshold absorbed fluence for P-to-AP switching is observed in all the studied samples, suggesting that the two types of switching are governed by different processes. This point will be discussed later. We also evaluate the threshold fluence by shining a laser pulse from the substrate side. In this case, a full P-to-AP switching is only observed for $t_{\text{Cu}} = 10$ nm (see Supplementary Information S3). Note that we carefully confirmed reproducibility in different

samples with same structure (10 nm Cu), which are deposited on a different date. We show the list of the studied samples in Supplementary Information S4.

In order to investigate the reversal speed, we measured the switching dynamics in the spin valve with $t_{\text{Cu}} = 10$ and 80 nm (see Methods for the definition of the reversal speed). Because the threshold fluence for AP-to-P switching is close to the one for the generation of the multidomain state, we prefer focusing on the P-to-AP switching. We pumped and probed the sample from the capping layer side (see Methods for details). For $t_{\text{Cu}} = 10$ nm, we utilize a quarter-wave plate (QWP) to distinguish the signals from the free and reference layers^{22,23,27}. This sample is deposited on a sapphire substrate. We confirmed that the substrate does not change the threshold incident fluence (see Extended Data Fig. 4). Figure 3a shows the switching dynamics for $t_{\text{Cu}} = 10$ nm at different fluences. For $F = 3.7$ mJ/cm², both the free (black filled symbol) and the reference layer (green empty symbol) are partially demagnetized before subsequently recovering. Increasing the fluence to $F = 6.1$ mJ/cm², the free layer (red filled symbol) is first demagnetized before recovering to the opposite direction within less than a picosecond. On the other hand, the reference layer (purple empty symbol) is not fully demagnetized (~70%) and its magnetization relaxes back to its initial state on a longer timescale. For $F = 11.1$ mJ/cm², both the free (blue filled symbol) and the reference layer (dark yellow empty symbol) reach full demagnetization leading to a complete demagnetization of both layers and no reversal of the free layer on a longer timescale.

Figure 3b shows the magnetization dynamics for $t_{\text{Cu}} = 80$ nm starting from P (green filled symbol) and AP (purple empty symbol) states. As the laser probe cannot reach the reference layer due to the 80 nm of Cu, only the magnetization dynamics of the free layer are observed. We find that the free layer magnetization switching occurs when starting from a P state, while it relaxes back to its initial state after ultra-fast demagnetization when starting from an AP state. This is consistent with the single-shot measurement shown in Fig. 2d. The magnetization

amplitude at a longer time scale is different between both samples, which may be due to the different heat dissipation between sapphire ($t_{\text{Cu}} = 10$ nm) and glass ($t_{\text{Cu}} = 80$ nm) substrates.

We have shown a single laser pulse-induced magnetization reversal of FM layer switching in rare-earth-free spin valves. Here we discuss the mechanism governing this magnetization reversal.

As already confirmed experimentally, when ultrafast demagnetization occurs, angular momentum conservation implies that the angular momentum will either dissipate locally into the lattice²⁸⁻³⁰ or will be transferred in the form of a spin current³¹⁻³⁵ to another part of the sample. Single pulse AOS in multilayers (even alloys) is understood in terms of mutual angular momentum exchanges between the magnetic sub-systems during their demagnetization^{13,15,18,20}. This tends to point towards the necessity of having antiparallel magnetic alignment of the sub-systems magnetization in order to observe any switching.

Here, surprisingly, we observe the opposite: a full switching of the free layer when starting from a P state. Simple angular momentum conservation arguments cannot explain our observation. We can also discard a common group of mechanisms as the source of the reversal, namely, magnetostatic couplings between the layers. Interlayer exchange can be eliminated due to the lack of exchange bias in the minor loops (shown in Extended Data Fig. 5) and because the reversal can still happen for the thick Cu sample. We also rule out dipolar couplings due to the following reasons: (i) No switching is observed when removing the reference layer (see Extended Data Fig. 6), which excludes dipolar (demagnetizing) fields from the free layer itself, (ii) the switching is independent of the shape of the surrounding domain structure (see Supplementary Information S2), which supports the previous point, (iii) the precessional half-period ($1/2\gamma M_S > 10$ ps, where γ is the electronic gyromagnetic ratio) due to a maximum dipolar field of the order of M_S (< 2 T) is much longer than the observed switching time, (iv) a faster reversal could be induced if the free layer reached its Curie temperature (T_C) ($M_S = 0$)

after excitation¹⁹, but the recovery of magnetization under a dipolar field would then be determined by the cooling of the magnetic bath and would still suffer from a critical slowing down^{28,36-39} of the dynamics around $M_S = 0$ which is not observed, and finally (v) we observe no P-to-AP switching for laser pulses with a pulse duration ≥ 3 ps (shown in Extended Data Fig. 7 and Supplementary Information S4), which indicates that the reversal is dominated by fast transients, and not by long-lasting magnetostatic interactions.

The reversal must therefore be due to some dynamical long-range coupling. We attribute such coupling to spin currents, which result in the injection of an opposite spin density (minority spins) into the free layer. However, the origin of such spin currents is not straightforward. Here we consider the following two scenarios: (i) in analogy to STT, the minority spins could originate from the free layer itself and be back-reflected at the interface with the reference layer (see left-hand side of Fig. 1c) or else (ii) the reference layer could generate a spin current of minority spins that is directly injected into the free layer (see right-hand side of Fig. 1c). In scenario (i), we include any mechanism that can inject minority spin carriers into the Cu layer from the free layer, which could then be reflected by the reference layer. For instance, these could include transient spin-polarized currents due to hot carrier excitation⁴⁰⁻⁴². We, therefore, propose a reflection mechanism (on the left-hand side of Fig. 1c), where the majority spins leaving the free layer are absorbed by the reference layer while the minority spins are reflected at the reference layer interface back to the free layer, resulting in P-to-AP switching. This mechanism is similar to the one observed in current-induced STT switching⁷. However, we note that in order to predict the angular momentum transfers in our optically excited systems one would need to account for both hot and cold electrons and their respective scattering probabilities^{43,44}. Concerning scenario (ii), we see no simple reason for the spin current to have an opposite polarity to that of the spins contained in the reference layer during its ultrafast demagnetization. However, at longer timescales, opposite spin currents have been observed. For instance, Choi *et al.*⁴⁵, showed that the SDSE in their [Co/Pt]/Cu structures induces spin currents

of opposite sign to the demagnetization-driven ones, albeit with a much smaller amplitude and on a much longer timescale, and therefore cannot explain our results. Also, it is known that after demagnetization, during the remagnetization ferromagnetic layers suck back via spin currents part of the spin they have lost, which has been confirmed by optical probing of spin accumulation^{34,45}. To compare the contributions from each mechanism, we calculate laser energy absorption profiles with different Cu thicknesses and irradiation directions as these mechanisms rely on the fact that either the free layer (scenario (i)) or the reference layer (scenario (ii)) is excited (demagnetized). Extended Data Figures 2c and 2d show the calculated energy absorption either in the free layer or the reference layer. As shown in Extended Data Fig. 2c, when the laser irradiates from the free layer, absorption in the free layer is almost constant with Cu thickness (blue dots), whereas the absorption in the reference layer drops to zero at around 40 nm (red dots). In our experiments with free layer side irradiation, we observe P-to-AP switching for all thicknesses (see shaded regions in Extended Data Fig. 2c). For an irradiation on the reference layer side, as shown in Extended Data Fig. 2d, the absorption in the reference layer has a small variation with Cu thickness (red dots), whereas the absorption in the free layer drops to zero at around 40 nm (red dots). Experimentally, we observe P-to-AP switching up to 20 nm Cu (see shaded regions in Extended Data Fig. 2d). These results tell us the following: exciting the free layer is enough for the P-to-AP switching (we do not need to excite the reference layer too much). Thus, we conclude that scenario (i) is a dominant driving force in the P-to-AP switching even though the detailed mechanism is unclear yet. Regarding scenario (ii), we performed simulations based on an *s-d* model⁴⁶, as shown in Fig. S7 (see Supplementary Information S6 for details). We could reproduce P-to-AP switching due to the remagnetization of the reference layer, but the reversal took place during the remagnetization of the reference layer (see Fig. S7). In contrast with the simulation results, the experiments show that the switching time of the free layer happens before the remagnetization of the reference layer (as shown in Extended Data Fig. 8), which means that, if present, the remagnetization mechanism is

most likely not dominant at short timescales. Moreover, this model relies on the excitation of the reference layer, which does not work with thick Cu when irradiated from the free layer. Thus, we conclude that scenario (ii) is not the driving force in the P-to-AP switching but could assist the P-to-AP switching after a few ps when Cu is thinner than 40 (20) nm when irradiated from the free (reference) layer, which allows excitation of the reference layer. Further studies are required to determine the exact cause of the minority spin currents. In all cases, we see an exciting parallel with STT switching, where P-to-AP switching is also possible.

At high fluences, we observe a (partial) AP-to-P reversal. We attribute this reversal to the spin current emitted by the reference layer during its demagnetization (Fig. 1b). This explains why the AP-to-P reversal seems to be more efficient when exciting the stack through the reference layer side. However, it is not clear yet why at low fluences the opposite spin injection process shown in Fig. 1c might dominate over the direct emission of spin from the reference layer (Fig. 1b). Our experiment and calculation of energy absorptions (Extended Data Fig. 2c and 2d) indicate that, for our samples, we should excite both the free layer and the reference layer for the AP-to-P switching unlike in the case of the P-to-AP switching. This could be one of the possible reasons for the dominance of P-to-AP switching at low fluences. In Supplementary Information S7., we discuss the difference between the two types of switching. Moreover, in Supplementary Information S8, we propose a proof of concept of bipolar switching by utilizing those two different switching mechanisms.

Finally, we compare the presented mechanisms to the conventional STT. As mentioned in the previous part, in conventional STT, a rapid increase in critical switching current with decreasing the pulse duration is unavoidable. Thus, low energy consumption is incompatible with high-speed switching below a few hundred picoseconds and large thermal stability (nonvolatility). On the other hand, with our optical excitations, the magnetization is temporarily demagnetized for an extremely short time (\sim picosecond), during which a magnetization reversal occurs due to spin transfer. Therefore, in principle, ultrafast magnetization reversal beyond a

few hundred picoseconds of highly anisotropic and non-volatile materials is possible, which challenges conventional STT. Since the studied spin valves are used in current-induced STT switching, future work will explore patterned devices to extend the playground of spintronics devices to ultrafast timescales.

In conclusion, we show that a single femtosecond laser pulse can induce the reversal of a Co/Pt ferromagnetic layer within a common spin-valve structure without any rare-earth element. We find two types of events depending on the laser fluence. The mechanism for P-to-AP switching is obviously different from the framework of current AOS and therefore fills a missing piece to achieve optical bipolar switching, as in current-induced STT and SOT switching. We believe this work provides a new route for ultrafast magnetization control by further bridging concepts from spintronics and ultrafast magnetism.

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Author contributions

J.I., M.H., G.M. and S.M. planned the study. M.H. performed sample fabrication and optimized the magnetic properties. J.I. and W.Z. performed single-shot measurements with input from G.M. J.I. and W.Z. performed layer-resolved TR-MOKE measurements with input from Q.R., J.G., G.M., and J.H. J.I., E.D., and J.G. performed TR-MOKE measurements without QWP. J.I., E.D., and J.G. performed the bipolar switching measurement. J.I. analyzed data with input from Q.R., J.H., J.G., and G.M. Q.R. performed simulations based on an *s-d* model. J.I. and J.G. wrote the manuscript with inputs from Q.R., E.D., J.X.L., M.H., S.M., and G.M. All authors contributed to discuss the measurement results.

Competing interests

The authors declare no competing interests.

Figure Captions

Figure 1 | Sample structure and schematic illustrations of the switching observed in this study. **a** Sample stack of spin valve consisting of the reference layer (bottom red layer), Cu spacer (middle yellow layer), and the free layer (top blue layer). Values in parentheses are layer thicknesses in nm. Schematic illustration of the process for magnetization reversal starting from **b** the antiparallel (AP) state and **c** the parallel (P) state based on the scenario (i) and (ii), explained in the discussion. Blue (red) spin-polarized electrons are generated in the free (reference) layer. Black arrow represents the direction of electron flow.

Figure 2 | MOKE hysteresis loops and results of magnetization switching using a single femtosecond laser pulse. **a** MOKE hysteresis loop measured from the reference layer for the sample with $t_{\text{Cu}} = 10$ nm. Lines in the graph show each magnetic state in the spin valve: P+ state (dark red line), AP+ state (blue line), AP- state (gray line), and P- state (gray line), respectively. **b** A sketch of the excitation (fs laser pulse) /probing (LED) geometries and MOKE images obtained after irradiation of a fs-laser pulse starting from the P+ (red) and AP+ (blue) states for $t_{\text{Cu}} = 10$ nm, with $F = 3.7$ and 5.9 mJ/cm². The color scale indicates the magnetic configurations. **c** MOKE hysteresis loop measured from the free layer side for the sample with $t_{\text{Cu}} = 80$ nm. Lines in the graph show each magnetic state in the spin valve: P+ state (dark red line), AP+ state (blue line), AP- state (gray line), and P- state (gray line), respectively. **d** A sketch of the excitation (fs laser pulse) /probing (LED) geometries and MOKE images obtained after

irradiation of a fs-laser pulse starting from the P+ (red) and AP+ (blue) states for $t_{Cu} = 80$ nm, with $F = 6.9$ and 12.0 mJ/cm². The black line in Fig. 2b and 2d is a 10 μ m scale bar.

Figure 3 | Magnetization dynamics obtained from TR-MOKE. **a** Magnetization dynamics for $t_{Cu} = 10$ nm measured for several fluences shown in the legend. Utilizing a QWP enables separating the signal from the free layer and from the reference layer. An external magnetic field larger than the coercive field of the reference layer is applied during the measurement. **b** Magnetization dynamics for $t_{Cu} = 80$ nm starting from the P+ (green) and AP+ (purple) states with $F = 10.4$ mJ/cm². An external magnetic field (H) whose magnitude lies between the coercive fields (H_C) of the free layer and the reference layer (H_C of free layer $< H < H_C$ of reference layer) is applied during the measurement.

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Methods

Film deposition. All samples were prepared by physical vapor deposition. The base pressure used to deposit multilayer film was 5×10^{-8} Torr. The basic stacking structures used in this study for the spin-valve structure were as follows:

Glass/Ta(5)/Pt(4)/[Co(1)/Pt(1)]₃/Co(1)/Cu(t_{Cu})/[Co(0.6)/Pt(1)]₂/Ta(5) (thickness in nm).

The Cu thicknesses (t_{Cu}) were 5, 10, 20, 40, 60, and 80 nm. Both [Co/Pt] multilayers show a strong perpendicular magnetic anisotropy (PMA). In this study, we observe magnetization switching in the free layer. The free layer has been designed to have the lowest Curie temperature.

For the TR-MOKE measurement with $t_{\text{Cu}} = 10$ nm, we used a spin-valve structure deposited on a sapphire substrate instead of glass. We confirmed no significant difference between these, at least regarding the threshold incident fluence for P-to-AP and AP-to-P switching (Extended Data Fig. 4).

Single-shot switching measurement. A Ti: sapphire fs-laser source and regenerative amplifier were used for the pump laser beam in the single-shot switching measurement. The wavelength, pulse duration, and repetition rate of the fs laser were 800 nm, ≈ 50 fs, and 5 kHz, respectively. The $1/e^2$ spot size $2w_0$ is ≈ 100 μm . MOKE images were obtained from the other side of the film. A LED light source with a wavelength of 628 nm was used for taking MOKE images.

We analyzed the obtained MOKE images in the same way as in Ref. ²⁰ for samples with $t_{\text{Cu}} = 5$ to 40 nm. For the threshold incident fluence of the AP-to-P switching, the position of the peak from the AP state is used (see Supplementary Information S1). We used the following equation

to extract the threshold incident fluence of each state as a fitting parameter: $r = w_0[1/2\ln\{E/(F_{th}\pi w_0^2)\}]^{0.5}$, where r , $F_{th,i}$, $2w_0$, and E are the domain radius, the threshold incident fluence, the $1/e^2$ spot size, and the energy of laser pulse, respectively. For samples with $t_{Cu} = 60$ and 80 nm, we analyzed the obtained MOKE images in the same way as in Ref. ¹⁵ due to the elliptical shape of the beam for an angle of incidence of 45° . We used the following equation to extract the threshold incident fluence of each state as a fitting parameter: Domain size = $\pi s w_0^2 [\ln\{E/(F_{th}\pi s w_0^2)\}]/2$, where s is the ratio between the long axis and short axis determined from MOKE images (~ 1.65 for both samples). We obtained the threshold absorbed fluence (Extended Data Fig. 3c) by multiplying $F_{th,i}$ (Extended Data Fig. 3a) and the calculated energy absorption in the free layer (Extended Data Fig. 3b).

Time-resolved MOKE measurement for $t_{Cu} = 10$ nm. The switching dynamics of this sample (Figure 3a) were measured using Ti: sapphire fs-laser source and regenerative amplifier to generate the linearly polarized pump (probe) laser beam with a central wavelength of 800 (400) nm. The repetition rate of the fs laser and pulse duration were 5 kHz and ≈ 50 fs, respectively. The pump and probe spot sizes were ≈ 200 μm and ≈ 50 μm , respectively. The measurement was performed with an out-of-plane magnetic field larger than the coercive field of both the free and the reference layer. A QWP was used to separate the signal of each layer^{22,23,27}.

Time-resolved MOKE measurement for $t_{Cu} = 80$ nm. The switching dynamics of this sample (Figure 3b) were measured using an amplified laser with a 10-kHz repetition rate and centered around 785 nm. The laser is split into two beams that become the pump and the probe. The pump and probe beams go through complementary high-pass and low-pass tint-filters⁴⁸ with a cutoff around ~ 780 nm, which allows to select part of the spectral profile of each of the beams. This operation avoids pump light from entering the balanced photodetectors, but results

in a widening of the pulse duration to ~ 300 fs. The pump beam passes through a delay line and is focused by a 15-cm lens before reaching the sample obliquely. The pump and probe spot sizes were ≈ 200 μm and ≈ 5 μm , respectively. The probe beam is modulated by a mechanical chopper at ~ 230 Hz and is focused through a 10x objective into the sample, resulting in a spot size of around 5 μm centered into the pump beam excitation area. The measurement was performed with an out-of-plane magnetic field of ~ 15 mT, which lies between the coercive field of the free layer and the reference layer.

The definition of magnetization reversal speed. We used the same definition for magnetization reversal speed with previous studies in the field of all-optical switching (see page 21 of Ref. ⁴⁹), which is the time delay at which the out-of-plane component of the magnetization crosses zero. We determined time zero (Time delay = 0) as the time delay where the derivative of magnetization dynamics is maximal. The magnetization when its out-of-plane component crosses zero is strongly demagnetized, and in order to recover its magnetization the magnetic layer needs to cool down and recover angular momentum. Such cooling normally happens on a much longer timescale but can be somewhat engineered via the use of heat sinks. Interestingly, it was recently shown that a 7 ps time delay between two subsequent laser pulses is enough time to allow cooling and re-toggle the magnetization via AOS⁵⁰, which means that recovering a full magnetization is not always necessary to operate ultrafast devices.

Calculation of the laser absorption profiles.

To estimate the energy absorption profiles in spin valves with different Cu thicknesses, we calculate light absorption using the transfer matrix method. The used parameters for the calculation are shown in Supporting Information in Ref. ⁴⁷. The energy absorption in the free (reference layer) is obtained by integrating absorbed energy density in the free (reference) layer.

Data Availability

The data that support the findings of this work are available from the corresponding authors upon reasonable request.

Code Availability

The code that calculated switching dynamics based on an s - d model is available from the corresponding authors upon reasonable request.

Methods-only references

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