Magnetization Reversal of a Ferromagnetic Pt/Co/Pt Film

by Helicity Dependent Absorption of Visible to Near-infrared Laser Pulses

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Abstract

The practical difficulty in distinguishing the impact of magnetic circular dichroism and the inverse Faraday effect fuels intense debates over which mechanism predominantly drives the process of helicity dependent all-optical switching of magnetization in ferromagnets. Here, we quantitatively measure the efficiency of the switching process in a Pt/Co/Pt multilayered stack using visible- to near-infrared optical pulses. We find that the switching efficiency increases by a factor of 8.6 upon increasing the pumping wavelength from 0.5 µm to 1.1 µm, becoming 100% efficient at even longer wavelengths up to 2.0 µm. Our experimental results can be successfully explained by the phenomenon of magnetic circular dichroism, making a significant step towards resolving the long-standing controversy over the origin of the all-optical process of magnetization reversal in ferromagnets.
The discovery of demagnetization on a sub-picosecond time scale by a femtosecond (fs) laser pulse gave birth to the new research field of ultrafast magnetism [1]. Furthermore, the use of circularly-polarized pulses allows magnetization dynamics to be selectively excited according to the optical helicity [2-4]. Helicity-dependent control of magnetization precession was first demonstrated in the dielectric DyFeO$_3$ by Kimel et al. [2], which was attributed to the Inverse Faraday effect (IFE) [3]. Subsequently, similar effects have also been observed in magnetic semiconductors [4] and metals [5,6]. For some magnetic metals, magnetization can be switched by fs circularly-polarized laser pulses in the absence of an external magnetic field. This phenomenon is referred to as all-optical helicity dependent switching (AO-HDS). AO-HDS was first demonstrated in ferrimagnetic GdFeCo [7] and later in ferromagnetic metals, including multilayered Co/Pt [8-15] and Co/Ni [10] stacks and granular FePt [16,17]. All-optical magnetization switching in GdFeCo films is driven by fast and efficient heating of electrons by the fs laser pulse [18] that brings the ferrimagnetic medium to a strongly non-equilibrium state [19]. The helicity dependence of the switching derives from magnetic circular dichroism (MCD) which leads to different laser absorption levels that depend on the helicity [20], but the switching, in general, relies only on the fast heating. In ferromagnetic metals, in contrast, it is evident that circularly polarized pulses are genuinely indispensable for AO-HDS. However, when discussing the dominant mechanism that causes the switching in such systems, one always encounters the outstanding question: do non-thermal or thermal effects drive the AO-HDS?

On one hand, the IFE is well known to be non-dissipative [3,21]. A circularly polarized fs pulse induces a magnetization in magnetic media via impulsive stimulated Raman processes [22], which is not accompanied by absorbing photons. Microscopic three-temperature models [23] and an
atomistic spin model [24] both show that the magnetic polarity of a ferromagnetic metal can indeed be switched by the IFE.

On the other hand, the absorption of a fs laser by a magnetic metal quasi-instantaneously elevates the metal’s temperature on a non-equilibrium timescale, resulting in ultrafast demagnetization [1]. Because the optical absorption of a ferromagnetic metal varies according to the helicity via the MCD, the helicity dependent laser absorption may be relevant to AO-HDS [24,25]. When magnetic domains of opposite polarity are illuminated by a circularly polarized pulse, their free energies decrease depending on the combination of optical helicity and magnetic orientation. As a result of repeating this process, magnetic domains with lower free energies should expand and eventually form a single magnetic domain, the final orientation of which depends on the helicity of the circularly polarized pulse.

In this Letter, we investigate multi-pulse AO-HDS of a ferromagnetic Pt/Co/Pt system in visible to near-infrared spectral ranges to elucidate whether AO-HDS is dominated by thermal or non-thermal effects. We experimentally find that the switching efficiency – defined as the amount of magnetization that is selectively switched by circularly-polarized light - is enhanced by a factor of 8.6 upon increasing the optical wavelength from 0.5 μm to 1.2 μm. A phenomenological model of the inverse Faraday effect cannot explain the improved switching efficiency. Instead we find that the effective magnetic field originating from the helicity dependent laser absorption increases by a factor of 32 in this spectral range, convincingly demonstrating that our results can be quantitatively described as a consequence of MCD.

Perpendicularly-magnetized Pt/Co/Pt structures have represented the most standard systems for studies of AO-HDS [8-15]. We therefore used similar materials, depositing Ta (1.0 nm)/MgO (2.0 nm)/Pt (3.0 nm)/Co (0.6 nm)/Pt (3.0 nm)/Ta (4.0 nm) on a synthetic glass
substrate by magnetron sputtering. We summarize in Fig. 1 the Faraday rotation $\theta_F$ and ellipticity $\eta_F$ of the Pt/Co/Pt stack in the range of wavelengths $\lambda$ between 0.5 $\mu$m and 1.2 $\mu$m, measured using a monochromator in combination with a photoelastic modulator [26]. Figure 1 shows that $\theta_F$ and $\eta_F$ increase, within this spectral range, by factors of ~3.0 and ~2.0 respectively. We also determine the magneto-optical parts of the refractive indices of the Co and Pt layers using a transfer matrix method (see Ref. [27] and Supplemental Note 1 [28] for details of this method). We thus calculate the MCD defined as $\delta_{\text{MCD}} = (A^- - A^+) / [(1/2)(A^+ + A^-)]$, where $A^{+(-)}$ is the total absorption of right-(left-) circularly-polarized light by the up-magnetized film. The spectral dependence of $\delta_{\text{MCD}}$ is also plotted in Fig. 1, showing that $\delta_{\text{MCD}}$ increases by a factor of ~4.2 with increasing $\lambda$ in the considered spectral range.

To excite the Pt/Co/Pt system, we used optical pulses ($\lambda = 800$ nm) delivered by an amplified Ti:Sa laser system at a repetition rate of 1 kHz. For $\lambda = 800$ nm, the pulse width was characterized using an autocorrelator to be 60 fs. By pumping an optical parametric amplifier (OPA), the central wavelength can be adjusted in the range $\lambda = 0.5 - 2.0$ $\mu$m. To obtain circular polarization across this broad spectral range, we used suitable quarter-wave plates AQWP05M-600 and AQWP05M-980 (Thorlabs Inc.) for $\lambda = 0.5 - 0.8$ $\mu$m and 0.95 – 1.1 $\mu$m, respectively, and PO-TWP-L4-25-IR (ALPHALAS GmbH) for $\lambda = 1.2 – 2.0$ $\mu$m. The train of circularly-polarized optical pulses was focused on the surface of the uniformly-magnetized Pt/Co/Pt sample. We characterized the Gaussian spot size on the sample at each wavelength by Liu’s method [20,29]. The incident fluence was determined by using the extracted spot size and the laser power measured in front of the sample. To evaluate the switching efficiency of AO-HDS, we perform sweeping experiments whereby the sample is mounted on a motorized stage, enabling the laser pulses to be swept 100 $\mu$m across the sample at a constant speed of 10 $\mu$m/s. We used the magneto-optical Faraday effect to
directly visualize the magnetization after exposure to the multiple optical pulses. Linearly-polarized white light illuminates the sample and is collected by a ×20 objective lens. Depending on whether the magnetization of the sample is parallel or antiparallel to the wave vector of the transmitted light, the latter’s polarization rotates in different senses. An analyzer, therefore, enables an image of magnetization to be detected using a charge-coupled device camera.

Figure 2(a) displays typical snapshots recorded after sweeping the Pt/Co/Pt sample with pulses at \( \lambda = 0.5 \, \mu\text{m}, 0.8 \, \mu\text{m}, 1.1 \, \mu\text{m} \) and \( 2.0 \, \mu\text{m} \) for the four cases \((M^\uparrow, \sigma^+), (M^\uparrow, \sigma^-), (M^\downarrow, \sigma^+)\) and \((M^\downarrow, \sigma^-)\) respectively. Here, we use the notation \((M^{\uparrow\downarrow}, \sigma^\pm)\) to indicate that the sample had initial magnetization pointing up \( M^\uparrow \) or down \( M^\downarrow \) and the impinging optical pulses were circularly-polarized with right-handed \( \sigma^+ \) or left-handed \( \sigma^- \) helicity. Figure 2(a) clearly shows that AO-HDS appears to become more efficient with increasing \( \lambda \). To quantify the spectral dependence of the AO-HDS, we estimate the net switched magnetization \( <M> \) as follows. First, we averaged the intensities in a central rectangular area spanning \( 80 \, \mu\text{m} \times 20 \, \mu\text{m} \) (364 × 91 pixels) on the track of the laser spot as shown in Fig. 2(a). Second, \( <M> \) was estimated by normalizing the average with a reference image recorded for the sample in a uniformly-magnetized state. Figure 2(b) shows \( <M> \) as a function of the laser incident fluence \( F \) for \( \lambda = 0.5 \, \mu\text{m}, 0.8 \, \mu\text{m}, 1.1 \, \mu\text{m}, \) and \( 2.0 \, \mu\text{m} \). The helicity dependence of the switching becomes increasingly stronger with increasing \( \lambda \). In particular, the helicity dependence emerges when the fluence exceeds the demagnetization threshold [Fig. S2], where switched domains start to appear at the center of the spot because the laser fluence is high enough to excite the magnetization beyond the Curie temperature. As previously reported [8], AO-HDS demands heating of the system close to the Curie temperature. Figure 3(a) plots the extracted switching efficiency as \( \varepsilon = [<M> (M^\uparrow, \sigma^-) - <M> (M^\uparrow, \sigma^+) + <M> (M^\downarrow, \sigma^+) - <M> (M^\downarrow, \sigma^-)]/4 \) versus \( \lambda \), where we average the net magnetization \( <M> \) obtained for all fluences larger than that
defining the demagnetization threshold. The spectral dependence of \( \varepsilon \) represents our key experimental finding and will be used later for quantitatively discussing which mechanism - the IFE or the MCD effect - principally drives the AO-HDS. The switching efficiency \( \varepsilon \) increases by a factor of \( \sim 8.6 \) from \( \lambda = 0.5 \, \mu m \) to \( \lambda = 1.1 \, \mu m \); \( \varepsilon \) reaches 100 \% (i.e., full switching) when \( \lambda \geq 1.2 \, \mu m \).

The pulse width is known to have a significant influence on AO-HDS. Extending the pulse durations to several picoseconds when optically irradiating multilayered [Pt/Co] \(_3\) stacks, for example, significantly promotes the switching efficiency compared to using 60-fs pulses [11]. It is therefore important to assess the pulse durations delivered across the broad range of wavelengths supplied by the OPA. We indirectly assessed the pulse width of the OPA output using single-shot all-optical switching in GdFeCo [30,31] (see the Supplemental Note 3 [28]). Fig S3 shows that the pulse width is nearly constant at \( \sim 100 \, fs \) for all wavelengths tested, except for an anomaly around 0.55-0.75 \( \mu m \) where it rises to 200-350 fs. Therefore, the observed improvement of \( \varepsilon \) cannot be explained by an extension of the pulse width with increasing \( \lambda \).

We first evaluate the spectral dependence of AO-HDS originating from the helicity dependent laser absorption via MCD. In general, this model [9,11,14] explains AO-HDS in terms of a two-step process. In the first step, the thermal load delivered by an optical pulse induces demagnetization in the central area of the Gaussian spot, creating many domain-walls (DWs) within a multi-domain state. In the second step, subsequent circularly-polarized optical pulses drive DW motion. The direction of the latter is defined by the MCD, since the circularly-polarized optical pulse is preferentially absorbed by one of the magnetic domains compared to the other. Because the ‘hotter’ domain has smaller free energy than the cooler domain, the DW moves opposite to the direction of the heat current [11]. Thus, the temperature gradient induced by the
difference in optical absorption (originating from MCD) leads to DW motion that ultimately switches the net magnetization.

To model this process, a DW excited by a circularly polarized pulse experiences an effective magnetic field $B_{\text{eff}}$ created by a one-dimensional thermal gradient $\nabla T$ [32]:

$$B_{\text{eff}} = -\frac{2}{M_s l} \frac{\partial A}{\partial T} \nabla T,$$

where $M_s = 1.13 \times 10^6 \text{ A/m}$, $A_0 = 1.6 \times 10^{-11} \text{ J/m}$ and $l_0 = 6.6 \text{ nm}$ denote the saturation magnetization, exchange stiffness and domain wall width respectively [30]. Assuming that the irradiated region is heated from room temperature ($T_0 = 300 \text{ K}$) towards the Curie temperature ($T_c = 470 \text{ K}$) [33], the thermal gradient acting across the DW is $\nabla T = \delta_{\text{MCD}} (T_c - T_0)/l_0$. Note that the thermal gradient is on the order of $0.01$-$0.1 \text{ K/nm}$, which is four orders of magnitude larger than that required to move the DW of a magnetic garnet [34]. Furthermore, we estimate the effective magnetic field $B_{\text{eff}}$ [Fig. 2(b)] associated with MCD assuming the temperature dependence of these magnetic parameters [35] as follows: $M(T) = M_0[1 - (T/T_C)^{2.37}]^{0.34}$, $A(T) = A_0[M(T) / M(0)]^{1.82}$, $l(T) = l_0[M(T) / M(0)]^{-0.59}$, and $\delta_{\text{MCD}} (T) = \delta_{\text{MCD}} (0)[M(T) / M(0)]$. Figure 3(b) shows that the estimated $B_{\text{eff}}$ arising from MCD increases by a factor of $\sim 32$ when increasing $\lambda$ from $0.5 \text{ \mu m}$ to $1.1 \text{ \mu m}$.

We next estimate the $B_{\text{eff}}$ produced by the IFE using a phenomenological approach [21]. The IFE induces a magnetization along the direction given by the cross product of the light’s electrical field and its complex conjugation, $\mathbf{E} \times \mathbf{E}^*$. The resultant magnetization, which is qualitatively equivalent to an effective magnetic field, is therefore nominally capable of switching the magnetization direction. In an isotropic magnetic medium, the strength of the effective magnetic field of the IFE [21] is given by
where $M_s, c, \tau, d,$ and $F$ denote the saturation magnetization, the speed of light, the pulse width, the stack thickness, and the optical fluence respectively. For the estimation, we fixed the pulse width at 60 fs for all wavelengths and used the thickness-weighted average of the real parts of the refractive indices of the Co and Pt layers [27]. Here, we considered the sum of the fluences, at the threshold of demagnetization, between an optical pulse propagating downward from the MgO/Pt interface and one propagating upward from the Pt/Ta interface. Although the handedness of a circularly polarized pulse changes by reflection, the IFE of the reflected pulse has the same sign as that of the incident pulse because of the inversion of the $k$ vector. Figure 3(b) indicates that the calculated effective magnetic field induced by the IFE is rather spectrally-invariant, with a maximum increase by a factor of $\sim 2.6$ at $\lambda = 0.8 \mu m$. This is in stark contrast to the clear experimental observation of $\varepsilon$ scaling monotonically with increasing $\lambda$ [Fig. 3(a)].

In the spectral range of 0.5 $\mu m$ – 1.1 $\mu m$, the switching efficiency $\varepsilon$ increases by a factor of $\sim 8.6$ [Fig. 3(a)]. In the same spectral range, the $B_{\text{eff}}$ from the MCD effect increases monotonically by a factor of $\sim 32$ while the $B_{\text{eff}}$ from the IFE only increases (non-monotonically) by $\sim 2.6$ [Fig. 3(b)]. The dramatic improvement of the AO-HDS with increasing $\lambda$ is quantitatively explained not by the spectral dependence of the IFE but rather by that of the MCD effect. Therefore, we conclude that the MCD effect predominantly drives the AO-HDS.

For $\lambda = 0.5 \mu m$, the switching efficiency $\varepsilon$ gradually elevates with increasing $F$ (see Fig. S4 [28]). The gradual increase of $\varepsilon$ might be due to an increased contribution from optical spin-orbit torques [36]. Recent studies [36] in particular have highlighted the ability of the latter to excite the magnetization dynamics in ferromagnetic multilayers, including Co/Pt. Ab-initio calculations may
be useful to more accurately discuss the contribution of the IFE to the AO-HDS since it is anticipated from equation (2) and the ab-initio calculations [37] that $B_{\text{eff}}$ from the IFE diverges for increasing $\lambda$ with a constant fluence. Investigating AO-HDS with infrared pulses in a free-electron laser facility [38,39] is a future work planned to gain more insight into the possibility of using IFE to achieve AO-HDS.

To summarize, we have investigated the spectral dependence of AO-HDS in a Pt/Co/Pt multilayered stack, in the visible to the near-infrared spectral regime, to ascertain the dominant mechanism that drives the process. We have experimentally found that the multi-pulse AO-HDS effect becomes increasingly efficient upon increasing the excitation wavelength, reaching 100% at wavelengths greater than 1.2 $\mu$m. The monotonic enhancement of the switching efficiency follows that of the effective field associated with the helicity dependent laser absorption, while the inverse Faraday effect cannot explain the improved AO-HDS. Our results provide crucial and experimentally-grounded insight into the origin of AO-HDS in ferromagnetic metals.

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FIG. 1. The experimentally-measured spectral dependences of Faraday rotation $\theta_F$, Faraday ellipticity $\eta_F$ and magnetic circular dichroism $\delta_{\text{MCD}}$ of the studied Pt/Co/Pt multilayered stack.
FIG. 2. (a) Typical results of multi-pulse all-optical helicity dependent switching (AO-HDS) for pumping wavelengths $\lambda$ of 0.5 $\mu$m, 0.8 $\mu$m, 1.1 $\mu$m, and 2.0 $\mu$m as indicated. Dark and bright contrasts correspond to up- and down-magnetized domains (pointing into or out of the plane of the page) respectively. Incident optical fluences ($F$) are shown below the snapshots. The scale bars correspond to 30 $\mu$m. We integrated the intensity in the rectangular of 20 $\mu$m × 80 $\mu$m to evaluate the net magnetization $<M>$. (b) $<M>$ as a function of $F$ for $\lambda = 0.5$ $\mu$m, 0.8 $\mu$m, 1.1 $\mu$m, and 2.0 $\mu$m. The red (blue) points corresponds to the case when the spin angular momentum of light is antiparallel (parallel) to the spin angular momentum of the original state.
FIG. 3. (a) The spectral dependence of the switching efficiency $\varepsilon$, obtained by averaging the efficiencies measured for varying incident fluences above the threshold of demagnetization. The solid line is a linear fit for points between $\lambda = 0.5 \mu m$ and $1.1 \mu m$. (b) The spectral dependencies of the effective fields $B_{\text{eff}}$ associated with the inverse Faraday effect (blue triangles) and the MCD effect (red squares). Note both vertical scales are adjusted to facilitate direct comparison of the spectral dependences. Inset: Unscaled spectral dependence of $B_{\text{eff}}$ associated with the inverse Faraday effect.
Supplemental Note 1: Estimation of the magneto-optical part of the refractive index based on a transfer-matrix formalism.

To calculate the amplitude of the light electric field at every interface, one must consider not only the light reflection and transmission of light at every interface between two different layers but also the absorption of light. Therefore, we used a well-known transfer matrix formalism [S1] to obtain the electric fields inside our multilayer by considering the refractive indices, $\tilde{n} = n + kj$, of the employed layers (Table S1). For normal incidence to a multilayer as depicted in Fig. S1, the Fresnel coefficients for the $m$-th interface are common for s- and p-polarized light:

$$r_{m,m+1} = \frac{n_m - n_{m+1}}{n_m + n_{m+1}}, \quad t_{m,m+1} = \frac{n_m}{n_m + n_{m+1}}. \quad (1)$$

Using the Fresnel coefficients, the electric fields on the left-hand and right-hand sides of the $m$-th interface are connected with an interface matrix $I_m$ as

$$
\begin{pmatrix}
E_{m}^{L\rightarrow} \\
E_{m}^{L\leftarrow}
\end{pmatrix} = \frac{1}{t_{m,m+1}} \begin{pmatrix}
1 & r_{m,m+1} \\
1 & 1
\end{pmatrix} \begin{pmatrix}
E_{m+1}^{R\rightarrow} \\
E_{m+1}^{R\leftarrow}
\end{pmatrix} \equiv I_m \begin{pmatrix}
E_{m}^{R\rightarrow} \\
E_{m}^{R\leftarrow}
\end{pmatrix}. \quad (2)
$$

The amplitudes and phases of the electric fields change as they travel across the $m$-th layer with a thickness $d_m$ and a refractive index $\tilde{n}_m$. The electric fields at the right-hand side of the $m$-th interface are connected with ones on the left-hand side of the $(m+1)$-th interface with a transport matrix $T_m$ according to

$$
\begin{pmatrix}
E_{m}^{R\rightarrow} \\
E_{m}^{R\leftarrow}
\end{pmatrix} = \begin{pmatrix}
\exp(-2\pi j \tilde{n}_m d_m / \lambda) & 0 \\
0 & \exp(2\pi j \tilde{n}_m d_m / \lambda)
\end{pmatrix} \begin{pmatrix}
E_{m+1}^{L\rightarrow} \\
E_{m+1}^{L\leftarrow}
\end{pmatrix} \equiv T_m \begin{pmatrix}
E_{m}^{L\rightarrow} \\
E_{m}^{L\leftarrow}
\end{pmatrix}. \quad (3)
$$
Therefore, the electric fields on the left side of the $m$-th interface is expressed with the electric fields at the left side of the $(m + 1)$-th interfaces as

$$\begin{pmatrix} E_{mL}^- \\ E_{mL}^+ \end{pmatrix} = I_m T_m \begin{pmatrix} E_{m+1L}^- \\ E_{m+1L}^+ \end{pmatrix}. \tag{4}$$

By repeating this process for all the interfaces and layers, one can obtain the relationship between the electric field amplitude of the incident ($i$), the reflected ($r$), and the transmitted ($t$) light waves:

$$\begin{pmatrix} i \\ r \end{pmatrix} = I_1 T_1 \cdots I_m T_m \cdots I_n T_n I_{n+1} \begin{pmatrix} T_0 \\ 0 \end{pmatrix} \equiv \begin{pmatrix} A_{11} & A_{12} \\ A_{21} & A_{22} \end{pmatrix} \begin{pmatrix} t \\ 0 \end{pmatrix}. \tag{5}$$

Solving this gives important relationships:

$$r = \left( \frac{A_{21}}{A_{11}} \right) i, \quad t = \left( \frac{1}{A_{11}} \right) i. \tag{6}$$

From energy conservation, the absorbed energy is

$$A = i^* i - r^* r - t^* t, \tag{6}$$

where ‘*’ denotes the complex conjugation. Also, the electric field amplitudes at a specific interface can be calculated with the interface and transport matrices; for instance, the electric field amplitudes at the $m$-th interface is

$$\begin{pmatrix} E_{mR}^- \\ E_{mR}^+ \end{pmatrix} = (I_1 T_1 \cdots I_{m-1} T_{m-1})^{-1} \begin{pmatrix} i \\ A_{21} i / A_{11} \end{pmatrix}. \tag{7}$$

The reflection changes the wave vector and the handedness of circularly polarized light. We note that applying the transfer matrix method to the magnetic layers requires consideration of not only the wave vector and handedness of circularly polarized light but also the magnetic orientations.
We estimated the refractive indices of the Co and Pt layers using the transfer matrix formalism for normal incidence. Throughout the estimation, for simplicity, we assumed that the magneto-optical part of \( \tilde{n} \) (i.e., \( n^+ - n^- \) and \( k^+ - k^- \)) is identical in the Pt/Co/Pt layer. The \( n^{+(-)} \), and \( k^{+(-)} \) denote the real and imaginary parts of the refractive index for right(left)-handed circularly polarized light, respectively. The assumption that the magneto-optical effect arises only from the Co layer gave unrealistically large values of \( n^+ \), \( n^- \), \( k^+ \), and \( k^- \). The Faraday rotation \( \theta_F \) is expressed as

\[
\theta_F = \frac{\pi d}{\lambda} (n^+ - n^-),
\]

where \( d \) is the total thickness of the Pt/Co/Pt multilayer. We determined the \( n^+ \) and \( n^- \) directly from the experimental value of \( \theta_F \). The Faraday ellipticity \( \eta_F \) is expressed as

\[
\eta_F = \tan^{-1} \left( \frac{|E^+| - |E^-|}{|E^+| + |E^-|} \right),
\]

where \( E^{+(-)} \) is the electric field of the transmitted right (left)-handed circularly polarized light.

Using the transfer-matrix method, we determined values of \( k^+ \) and \( k^- \) that match the experimental value of \( \eta_F \) for the up-magnetized case. Table S1 summarizes the estimated values of \( \tilde{n} \) of Co and Pt layers when the magnetization is antiparallel to the light wave vector. We moreover determined the magnetic circular dichroism [Fig. 1(c)] with the determined refractive indices based on equation (6).

**Supplemental Note 2: Demagnetization threshold.**

In the main text, we defined the demagnetization threshold as the optical fluence at which switched domains start appearing at the center of the laser spot as in Fig. S2(a). Figure S2(b) shows
the demagnetization threshold and the absorbed energy as a function of the wavelength. While the absorbed energy at the demagnetization threshold is almost constant up to 800 nm, it decreases by a factor of 4.2 with further increasing of the wavelength.

Supplemental Note 3: Assessing the wavelength-dependent pulse duration

The process of AO-HDS is well-known to depend on the duration of the optical pulses. Ref. S2 showed, for example, that 4-ps-long circularly polarized pulses (wavelength 800 nm) are much more efficient in switching magnetization compared to fs pulses. It is therefore important for us to evaluate the duration of the visible and near-infrared pulses outputted by the OPA. In lieu of suitable broadband autocorrelators, we indirectly estimate the pulse width by measuring the spectral and thermal dependence of single-shot all-optical switching in a GdFeCo film, which is susceptible to both the pulse width [S11] and starting temperature [S12].

To evaluate the pulse duration, we tested the ability of the optical pulses to achieve single-shot helicity-independent all-optical switching in a GdFeCo sample. Previous experiments have shown that this process depends not on the optical wavelength but rather on the pulse duration \( \tau \) [S11]. If \( \tau > \tau_c \) where \( \tau_c \) is a cutoff duration, the pulse demagnetizes the optically-exposed area, whereas \( \tau < \tau_c \) results in single-shot switching of magnetization. Moreover, \( \tau_c \) is linearly dependent on the starting temperature \( T_0 \) so that as \( T_0 \) increases, \( \tau_c \) decreases [S12]. This implies that for a fixed pulse duration, there is a cutoff starting temperature \( T_{\text{cutoff}} \) above which the switching is disabled (see Fig. S3(a)).

Using these results, we are able to assess the duration of the optical pulses outputted by the OPA. A 20 nm-thick sample of Gd\(_{23}\)(FeCo)\(_{77}\) is mounted on a resistive heater, and exposed to optical
pulses of varying wavelength as supplied by the OPA. Details of the experimental setup are otherwise the same as those given in Ref. S11.

For a fixed optical wavelength, $T_0$ is steadily increased until single-shot all-optical switching is no longer possible, i.e., we identify $T_{\text{cutoff}}$. This method is repeated for varying wavelengths, producing a dependence $T_{\text{cutoff}}(\lambda)$ as shown in Fig. S3(b). Next, by varying the duration of the 800 nm-pulses, we measure a state diagram for switching as a function of both $\tau$ and $T_0$. This is shown in Fig. S3(c). Linear fitting of the boundary between the two regions yields the dependence $\tau_c(T_0)$. Finally, by exploiting the known spectral indifference of $\tau_c$, we transform the cutoff starting temperature to the cutoff duration, thus obtaining the duration of the visible and near-infrared pulses as shown in Fig. S3(d).

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TABLE S1. The refractive indices used for the transfer-matrix analysis. For the transfer-matrix method, we used refractive indices of Co, Pt, Ta, MgO, Ta$_2$O$_5$, and glass in Ref. S5–S10, respectively.

| Central Wavelength $\lambda$ (nm) | Refractive index $\tilde{n} = n + kj$ |
|-----------------------------------|-----------------------------------|
| Ta$_2$O$_5$                       | MgO                               |
| 500                               | 2.159                             |
| 600                               | 2.128                             |
| 700                               | 2.109                             |
| 800                               | 2.096                             |
| 950                               | 2.083                             |
| 1025                              | 2.078                             |
| 1100                              | 2.074                             |
| Co                                | 1.746                             |
| 1.991 + 3.448j                    | 2.268 + 3.967j                    |
| 2.566 + 4.488j                    | 2.839 + 4.950j                    |
| 3.283 + 5.591j                    | 3.522 + 5.889j                    |
| 3.773 + 6.153j                    | 3.773 + 5.258j                    |
| Pt                                | 1.734                             |
| 2.268 + 4.202j                    | 2.566 + 4.480j                    |
| 2.839 + 4.972j                    | 3.283 + 4.998j                    |
| 3.522 + 5.128j                    | 0.982 + 1.878j                    |
| Ta                                | 1.731                             |
| 1.991 + 3.479j                    | 2.268 + 4.202j                    |
| 2.566 + 4.480j                    | 2.839 + 4.712j                    |
| 3.283 + 4.998j                    | 0.982 + 1.878j                    |
| glass                             | 2.019                             |
| 1.462                             | 1.458                             |
| 1.458                             | 1.453                             |
| 1.453                             | 1.451                             |
| 1.450                             | 1.449                             |

FIG S1. (a) A schematic of a thin multilayer with $n$ layers. (b) A schematic of the $m$-th layer with the light electric field amplitudes. The superscript letters and arrows denote the side of the interface and the light propagation direction, respectively. For instance, the notation $E_m^{L\to}$ corresponds to the electric field incident to the left side of the $m$-th interface.
TABLE S2. The magneto-optical parts of the refractive indices of the Co and Pt layers

| Central Wavelength λ (nm) | Refractive index \( M'\bar{n} = n + kj \) |
|---------------------------|------------------------------------------|
|                           | Co                                       | Pt                                       |
|                           | \( \sigma^+ \)                            | \( \sigma^- \)                           | \( \sigma^+ \)                            | \( \sigma^- \)                           |
| 500                       | 2.042 + 3.680j                           | 2.009 + 3.760j                           | 1.987 + 3.408j                           | 1.954 + 3.488j                           |
| 600                       | 2.064 + 4.132j                           | 2.589 + 4.272j                           | 2.281 + 3.897j                           | 2.230 + 4.037j                           |
| 700                       | 3.210 + 4.365j                           | 3.142 + 4.595j                           | 2.574 + 4.373j                           | 2.506 + 4.603j                           |
| 800                       | 3.671 + 4.529j                           | 3.565 + 4.895j                           | 2.892 + 4.767j                           | 2.786 + 5.133j                           |
| 950                       | 4.058 + 4.635j                           | 3.896 + 5.342j                           | 3.364 + 5.247j                           | 3.202 + 5.935j                           |
| 1025                      | 4.159 + 4.726j                           | 3.986 + 5.530j                           | 3.609 + 5.487j                           | 3.436 + 6.291j                           |
| 1100                      | 4.277 + 4.749j                           | 4.059 + 5.767j                           | 3.882 + 5.644j                           | 3.665 + 6.662j                           |
FIG S2. (a) A typical magneto-optical snapshot at the demagnetization threshold. The scale bar corresponds to 10 μm. (b) The spectral dependence of the demagnetization threshold. The absorbed fluences were calculated by considering the total light absorption by the multilayer.
FIG. S3. (a) Typical magneto-optical images recorded after exposing GdFeCo to consecutive optical pulses at $\lambda = 1.8 \, \mu m$. The images in the top row were measured with a sample temperature $T_0 = 388 \, K$, whereas in the bottom row, $T_0 = 295 \, K$. (b) State diagram showing whether switching (blue area) or demagnetization (red area) is achieved by pulses at $\lambda = 0.8 \, \mu m$, with the indicated pulse duration and $T_0$. (c) State diagram showing whether switching is successfully obtained with different $\lambda$ and $T_0$. (d) Estimated spectral dependence of the pulse duration outputted by the OPA, obtained using the results shown in panel (c) and the assumption that the cutoff temperature for switching is wavelength-independent.
FIG S4. The switching efficiency $\varepsilon$ as a function of the incident fluence at $\lambda = 500$ nm. The red line denotes the linear fitting.