Twisted light affects ultrafast demagnetization

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Abstract:
Irradiation with an ultrashort laser pulse can completely destroy the magnetic order of ferromagnetic thin films on the femtosecond timescale. This phenomenon holds great potential for ultrafast spintronics and information processing and is an active field of research. It is still an open question if the angular momentum of light can support this effect. While it has been shown that the spin of light only has a negligible influence, we experimentally demonstrate the influence of ultrashort laser pulses with orbital angular momentum (OAM) on the magnetization dynamics of a thin nickel film. Our results reveal that the photonic OAM affects the demagnetization behavior within the first hundreds of femtoseconds depending on the handedness of the OAM with respect to the direction of the sample magnetization.

Main Text:
Photons can carry two types of angular momentum. As bosons, they always have a spin $S = \pm \hbar$ (with the spin quantum number $s = \pm 1$ and the reduced Planck constant $\hbar$) that is associated with left- or right-handed circular polarization, respectively. In addition, they can also carry an orbital angular momentum (OAM) $L = l \hbar$ (with the OAM quantum number $l \in \mathbb{Z}$) that leads to characteristic annular beam profiles with a singularity at the center and a spiral energy flow. It has been shown that photonic OAM, often referred to as twisted light, can be transferred to matter to mechanically rotate micro-particles [4] as well as to influence higher-order electronic transitions [5]. Both types of angular momentum are used for various applications, such as data transmission [6,7], quantum information technology [8,9], and micromanipulations in optical tweezers [10,11].

Angular momentum is directly related to magnetic moments, the source of magnetism. Thus, the vision to control magnetization and its optically induced dynamics [12] with photonic OAM is very intuitive. In general, the ultrafast, femtosecond time scale magnetization dynamics is triggered by irradiation of magnetic samples with ultrashort laser pulses. Here, energy can be deposited within tens of femtoseconds onto the material while the conservation of angular momentum, which must be transferred from the electron spin to the lattice [13], is a limiting factor for the speed of the demagnetization. During this process, photonic angular momentum from the laser pulse could affect the angular momentum balance in the material. First attempts explored the influence of the light’s spin on the magnetization dynamics. The spin of light, however, has been shown to have no measurable influence on the ultrafast demagnetization of simple ferromagnetic thin films such as Ni [14,15], but it is crucial for the so-called helicity-dependent all-optical switching (HD-AOS) [16,17]. This phenomenon has been observed in ferromagnetic rare-earth transition-metal alloys as well as in ferromagnetic multilayer systems and is solely based on the spin degree of freedom of the photons, either through the inverse Faraday effect (IFE) [18,19] or through magnetic circular dichroism (MCD) [19–21].
In our work, we aim to gain new insights into whether photonic OAM can also be used to control magnetization on ultrafast timescales. Recent publications already demonstrated a dichroism effect for terahertz radiation with OAM in the rare earth iron garnet Dy$_3$Fe$_5$O$_{12}$ [22] and the possibility to modify the magnetic anisotropy of an interlayer exchange coupling system with OAM light [23]. In our study, we focus directly on the transfer of angular momentum into a magnetic material at the time scale of the optical excitation. To this end, we illuminated a thin nickel film with linearly polarized ultrashort laser pulses carrying varying orders of OAM. By recording the magnetic response of the sample, we observed that the OAM either supports or obstructs the demagnetization process, depending on the direction of the OAM vector with respect to the magnetization direction of the sample.

To investigate the influence of photonic OAM on ultrafast laser-induced demagnetization, we chose the longitudinal time-resolved magneto-optical Kerr effect (TR-MOKE) technique. The sample consists of a 10 nm thick polycrystalline nickel film deposited on a MgO substrate and is protected by a 100 nm Si$_2$O$_3$ capping layer. The sample was optically excited with ultrashort ($\tau_{\text{pump}} \leq 35$ fs), linearly polarized laser pulses from a Ti:Sa amplifier with a central wavelength of $\lambda_{\text{pump}} = 800$ nm carrying an OAM of $l \in [-8, 8]$. We probed the magnetic response of a position on the ring of the OAM mode (see the SM for beam camera images) with the linearly polarized second harmonic of the amplifier with a central wavelength of $\lambda_{\text{probe}} = 400$ nm ($\tau_{\text{probe}} \leq 75$ fs) that did not carry an OAM ($l_{\text{probe}} = 0$).

To obtain the intrinsic magnetic response in TR-MOKE experiments, it is common practice to perform the time-resolved measurement for opposite magnetization directions of the sample. The pure magnetic material response is then extracted by the difference of the two resulting curves. This excludes any signal contributions that are independent of the direction of the sample magnetization, such as, for example, the specular inverse Faraday effect (SIFE) and specular optical Kerr effect (SOKE) [24]. These nonmagnetic signal contributions can be obtained as the sum of the two curves.

However, to investigate and determine the influence of photonic OAM, the direction of the OAM vector $L$ with respect to the sample magnetization $M$ must also be taken into account. This leads to a total of four different measurement geometries. For two of them, the projection of the OAM vector onto the magnetization direction of the sample and the magnetization are parallel ($L \cdot M > 0$, see Fig. 1, A and B) and the light’s OAM supports the demagnetization process. For the other two geometries, the directions are antiparallel ($L \cdot M < 0$, Fig. 1, C and D), and the OAM obstructs the demagnetization. To reveal this influence of the photonic OAM, the differences of the supporting and obstructing cases must be considered, respectively (see the SM for details).
Figure 1: Schematic of the four different measurement geometries used to investigate the influence of optical OAM on laser-induced demagnetization. (A, B) Supporting geometries. In these geometries, the direction of the sample magnetization $M$ and the projection of the OAM vector $L$ (of the pump-pulse) into the sample plane are parallel. (C, D) Obstructing geometries. In these cases, $M$ and $L$ are antiparallel.

Figure 2 compares the magnetic responses of the sample measured and extracted with the method described above for irradiation of $|l| = 5$ (Fig. 2A) and $|l| = 7$ (Fig. 2B) with the response for $l = 0$ illumination (black). The supporting (blue) and obstructing (red) cases clearly show a different behavior from one another as well as from the $l = 0$ measurement. At a pump-probe delay of $\Delta t = 0$ fs, the drop of the magnetization begins similarly. However, with an increasing pulse delay, the curves split up with the supporting case demagnetizing more strongly than the obstructing case. The difference reaches its maximum at $\Delta t \approx 150$ fs. After this, the two curves reunite again and follow the same trace. Compared to the demagnetization curve without OAM, it looks like both the supporting and the obstructing curves have “bumps” in the area of separation that point in opposite directions. A similar behavior was observed for the other OAM orders (see the SM).
Figure 2: Comparison of laser-induced demagnetization with and without photonic OAM. To best visualize the influence of the OAM, the data points were smoothed (see the SM) and connected with lines. (A) Demagnetization of the sample after irradiation of a pump pulse with $|l| = 5$ in the supporting (blue) and obstructing (red) geometry in comparison to demagnetization with an $l = 0$ beam (black). (B) Demagnetization of the sample after irradiation of a pump pulse with $|l| = 7$ in the supporting and the obstructing geometry in comparison to demagnetization with an $l = 0$ beam. The purple curves in both plots visualize the respective pump-pulse durations during the $|l| = 5$ and $|l| = 7$ measurements.

To quantify the influence of photonic OAM on laser-induced ultrafast demagnetization of nickel, we developed a simple fitting model (see the SM for a more detailed description). The main focus of this model is to determine whether the OAM affects the magnetization instantaneously, as it would be the case for a primary effect, or with a delay, indicating the involvement of a secondary process. The model assumes that the supporting and obstructing cases share a common temporal evolution of the thermally-induced demagnetization similar to the demagnetization with $l = 0$ light and approximates this with a double exponential function (for the de- and remagnetization processes). To determine the influence of the photonic OAM, we model it with an additional Gaussian curve that is added to the double exponential function. We assume that this effect is, except for its sign, identical for the supporting and the obstructing cases. The total magnetic response is then convolved with a Gaussian that accounts for the experimental resolution due to the finite durations of the pump and probe pulse. Fig. 3 displays examples of experimental data with different OAM orders that were fitted with this model. The results clearly show that the effect of the OAM (the dark blue and dark red curves in Fig. 3) is delayed with respect to the beginning of the demagnetization, i.e. it evolves on timescales larger than the temporal width of the pump pulses.
Figure 3: Fitting Model. Fitting of laser-induced demagnetization for different orders of photonic OAM. (A) $|l| = 1$, (B) $|l| = 3$, (C) $|l| = 5$, and (D) $|l| = 7$. The light blue and light red dots represent the experimental data for the supporting and the obstructing case, respectively. The black lines correspond to the common demagnetization behavior, while the dark blue and red lines represent the additional Gaussians that are added to the shared behavior to account for the supporting/obstructing influence of the photonic OAM (with an offset of $+1$ for better visualization). The bright blue and red lines show the combinations of the shared behavior and the OAM influence.

In addition, our model allows us to determine how the magnetic response of the sample depends on the order of the incident OAM. For this, the most interesting parameter is the center position of the Gaussians that account for the OAM influence on the demagnetization (the dark blue and dark red lines in Fig. 3). This is the time at which the OAM contribution reaches its maximum. In Fig. 4, we plotted this value from all our measured datasets with different OAM orders (see the SM for all datasets that are not shown in Fig. 3) against the order of the incident OAM $|l|$.

Our results strongly suggest that the OAM contribution to the demagnetization process reaches its maximum faster with an increasing OAM order of the incident light, with a difference of more than 100 fs between $|l| = 1$ and $|l| = 8$. Crucially, all these center times are considerably larger than the duration of the pump pulse ($\tau_{pump} \leq 35$ fs). This clearly shows that the influence of the photonic OAM on the demagnetization dynamics must be caused by a secondary process.
Figure 4: Correlation between the OAM order and its influence on the demagnetization process. The data points in this plot were obtained from the fitting model shown in Fig. 3. They correspond to the time between the values for $\Delta t = 0$ fs and the centers of the Gaussians that simulate the OAM influence on the demagnetization for different OAM orders. The error bars are based on the fitting errors.

All these findings are different from the photonic spin, for which Koopmans et al. [14] demonstrated theoretically and Dalla-Longa et al. [15] demonstrated experimentally that there is neither a measurable primary [14,15] nor secondary [15] effect. Specifically, regarding a primary process, these studies have shown that the photon absorption probability per Nickel atom at a realistic pump fluence is too small for a relevant direct transfer of spin from the photons to the electrons, contributing at most $\pm 0.01\%$ to the demagnetization. Therefore, according to the calculations from Ref. [14], an OAM of more than $100\hbar$ per photon would be necessary for a measurable influence of the OAM of light in a primary process (assuming that it was possible to transfer this amount of angular momentum in a single absorption process). This fits well with our experimental results that do not show an instantaneous effect as it would be expected if the electronic transitions were influenced by a primary process like a direct transfer of angular momentum or dichroism. In the latter case, we would also expect either a stronger or weaker overall demagnetization for the supporting and obstructing geometries, respectively, instead of an effect that is delayed and decays after less than 500 fs. Instead, our observations suggest the involvement of secondary processes.

A possible example of such a secondary process is the induction of a circular current by the photonic OAM, which has been observed for the type-II Weyl semimetal WTe$_2$ [25] and theoretically predicted for semiconductors [26] as well as plasmas [27]. This phenomenon has also been discussed as a potential reason for the OAM light-induced change in the magnetic anisotropy of an interlayer exchange coupling system [23] with the current inducing a magnetic field. This is similar to the inverse Faraday effect (IFE), which refers to the induction of a magnetic field [28] or magnetization [29] by circularly polarized light and has also been theoretically studied for light with OAM, however, only for a plasma [30] and a thin gold film [31]. Alternatively, the photonic OAM might first be transferred to the electronic OAM before affecting the electronic spin.

In conclusion, we have demonstrated that the OAM of light has a significant influence on the ultrafast demagnetization dynamics of a thin nickel film. This effect is delayed with respect to the excitation of the system and can, therefore, be attributed to a secondary process. Depending on the measurement geometry, the OAM temporarily either supports or obstructs the demagnetization process for a time span of hundreds of femtoseconds. We also showed that the effect occurs more rapidly with an increasing OAM order. Our results could lead to new means of control for all-optical switching and manipulating spin-ordered systems.
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Supplementary Materials

Experimental setup

In our experiments, we used a Ti:sapphire amplifier (Dragon from KMLabs) to create ultrashort (~35 fs) laser pulses at a central wavelength of 800 nm with a repetition rate of 6 kHz. For the pump-probe measurements, each pulse was split into two pulses, which were delayed with respect to each other in 10 fs steps with a Mach-Zehnder interferometer. The probe beam was frequency-doubled with a β-barium borate (BBO) crystal to a central wavelength of 400 nm before being focused onto the sample with a pulse length of < 80 fs. The pump beam was sent through a spiral phase plate (VPP-m780 from RPC photonics) with the order 1 – 8, adding OAM to the pulses. Both beams were linearly polarized and their intensities were adjusted with combinations of half-wave plates and linear polarizers.

The TR-MOKE experiments were performed with the setup that is schematically depicted in Fig. S1. We used a longitudinal geometry with an angle of incidence of ~45° of both beams with respect to the sample normal. To monitor changes in the sample magnetization, we measured changes in the polarization of the probe beam after reflection from the sample. An external magnetic field was applied to saturate the sample. A flip mirror allowed us to monitor the overlap of the pump and probe beams with a beam camera in sample distance before each measurement.

Data evaluation procedure

For each data set, we performed four time-resolved measurements of the different combinations of sample magnetization direction and incident OAM handedness, respectively. While the sample magnetization could easily be changed by switching the direction of the external magnetic field, changing the OAM handedness required turning the spiral phase plate by 180°. Since the latter made it necessary to readjust the beam overlap, this could lead to a change in time zero. To account for this, we compared the sum signals of the two measurements with the same OAM handedness, respectively, which correspond to nonmagnetic signal contributions, and matched them by shifting the data corresponding to one OAM handedness in time. Figs. S2-S14A show the (normalized) resulting data.

The measurements with different OAM handedness can only be used for further evaluation if they were performed with similar fluences in the probed regions. Before the measurements, we optimized this with the beam camera (see insets in Figs. S2-S14D, the brightness of the images can vary due to a grey filter wheel that had to be manually adjusted before each measurement) and during the evaluation, we verified the quality of our adjustment by calculating and comparing the sum signal of the supporting and obstructing excitation geometries, respectively. These nonmagnetic signal contributions (Figs. S2-S14B) should ideally be identical.

To see if the photonic OAM influences the demagnetization behavior, we calculated the difference signal of the supporting and obstructing geometries, respectively (see Figs. S2-S14C).

Figures S2-S14D show the sum of the magnetic signals of the supporting and obstructing geometries. The resulting curves resemble “normal” demagnetization curves from pumping
light without OAM, indicating that the supporting and obstructing OAM influences are of similar magnitude as they cancel each other out.

Fitting model

Our fitting model for the ultrafast demagnetization with OAM light is composed of three different contributions:

1. The “normal” demagnetization without the OAM influence with a single exponential decay followed by an exponential increase:

   \[ s_{demag}(\Delta t) = M_0 \cdot (-\Theta(\Delta t - t_0) + 1) \]
   \[ + \left( M_{relax} + (M_0 - M_{min}) \cdot e^{\Delta t-t_0 \over \tau_{demag}} \right) \]
   \[ + (M_{min} - M_{relax}) \cdot e^{\Delta t-t_0 \over \tau_{relax}} \cdot \Theta(\Delta t - t_0) \]  

   with \( \Delta t \) the time delay between pump and probe pulse, \( M_0 = 1 \) the initial (saturation) magnetization, \( \Theta \) the Heavyside step function, \( t_0 \) the temporal overlap of pump and probe pulse (time zero), \( M_{min} \) the magnetization to which the exponential decay converges, \( \tau_{demag} \) the time constant of the exponential decay, \( M_{relax} \) the magnetization to which the exponential increase converges, and \( \tau_{relax} \) the time constant of the exponential increase.

2. The influence of the photonic OAM on the demagnetization process which we model with a Gaussian:

   \[ s_{OAM}(\Delta t) = \left( \frac{S}{\sqrt{2\pi}} \cdot \frac{\sqrt{2 \ln(2)}}{\sigma_{OAM}} \cdot e^{-(\Delta t-t_{center})^2(2\sqrt{2\ln(2)}^2)} \right) \cdot \Theta(\Delta t - t_0) \]  

   with \( S \) a scale factor, \( \sigma_{OAM} \) the FWHM, and \( t_{center} \) the center position of the Gaussian.

3. The experimental time smear due to the finite lengths of pump and probe pulse:

   \[ s_{time\_smear}(\Delta t) = \frac{1}{\sqrt{2\pi}} \cdot \frac{\sqrt{2\ln(2)}}{\sigma} \cdot e^{-\Delta t^2(2\sqrt{2\ln(2)}^2)} \]  

   with \( \sigma = 83 \) fs the FWHM. This value was determined experimentally from an XFROG trace of a pump pulse without OAM and a probe pulse. \( s_{time\_smear} \) is normalized to \( s_{time\_smear,norm} \) for all further calculations.

For fitting the supporting contributions, we calculate the difference

\[ s_{sup}(\Delta t) = s_{demag}(\Delta t) - s_{OAM}(\Delta t), \]
while for the obstructing contributions, we calculate the sum

$$s_{obs}(\Delta t) = s_{demag}(\Delta t) + s_{OAM}(\Delta t).$$  \hspace{1cm} (S5)

To account for the time smear, we convolve both functions with $s_{time\_smear}(\Delta t)$, respectively:

$$s_{sup/obs\_time\_smear}(\Delta t) = s_{sup/obs}(\Delta t) \circ s_{time\_smear, norm}(\Delta t).$$  \hspace{1cm} (S6)

Finally, we simultaneously fit $s_{sup\_time\_smear}(\Delta t)$ and $s_{obs\_time\_smear}(\Delta t)$ with the same fitting parameters. These functions correspond to the blue and red curves in Fig. 3 and Fig. S10, respectively. The black curves in these figures are given by $s_{demag}(\Delta t) \circ s_{time\_smear, norm}(\Delta t)$ while the dark blue and dark red curves are given by $1 + s_{OAM}(\Delta t) \circ s_{time\_smear, norm}(\Delta t)$ and $1 - s_{OAM}(\Delta t) \circ s_{time\_smear, norm}(\Delta t)$, respectively.

The data with $l = 0$ we fitted with

$$s_{l=0}(\Delta t) = s_{demag}(\Delta t)s_{time\_smear, norm}(\Delta t).$$  \hspace{1cm} (S6)
Fig. S1.
Schematic of the longitudinal TR-MOKE setup.
Fig. S2.

Experimental data of $|l| = 1$. For optimal visualization, the data points were connected with lines. The blue lines correspond to data from supporting experimental geometries while the red curves correspond to data from obstructing geometries. (A) Normalized and shifted (in time) experimental data from the four possible excitation geometries. (B) Nonmagnetic sum signal, calculated from the supporting and obstructing data in (A), respectively. (C) Magnetic difference signal, calculated from the supporting and obstructing data in (A), respectively. (D) Sum of the supporting and obstructing signals in (C). The insets are beam camera images that show the pump-probe overlap for $l = 1$ (left) and $l = -1$ (right). The scale bars are 1 mm.
Fig. S3.
Experimental data of $|l| = 2$. For optimal visualization, the data points were connected with lines. The blue lines correspond to data from supporting experimental geometries while the red curves correspond to data from obstructing geometries. (A) Normalized and shifted (in time) experimental data from the four possible excitation geometries. (B) Nonmagnetic sum signal, calculated from the supporting and obstructing data in (A), respectively. (C) Magnetic difference signal, calculated from the supporting and obstructing data in (A), respectively. (D) Sum of the supporting and obstructing signals in (C). The insets are beam camera images that show the pump-probe overlap for $l = 2$ (left) and $l = -2$ (right). The scale bars are 1 mm.
Fig. S4.

Experimental data of $|l| = 3$ with a lower pump fluence than the data in Fig. S5. For optimal visualization, the data points were connected with lines. The blue lines correspond to data from supporting experimental geometries while the red curves correspond to data from obstructing geometries. (A) Normalized and shifted (in time) experimental data from the four possible excitation geometries. (B) Nonmagnetic sum signal, calculated from the supporting and obstructing data in (A), respectively. (C) Magnetic difference signal, calculated from the supporting and obstructing data in (A), respectively. (D) Sum of the supporting and obstructing signals in (C). The insets are beam camera images that show the pump-probe overlap for $l = 3$ (left) and $l = −3$ (right). The scale bars are 1 mm.
Fig. S5.

Experimental data of $|l| = 3$ with a higher pump fluence than the data in Fig. S4. For optimal visualization, the data points were connected with lines. The blue lines correspond to data from supporting experimental geometries while the red curves correspond to data from obstructing geometries. (A) Normalized and shifted (in time) experimental data from the four possible excitation geometries. (B) Nonmagnetic sum signal, calculated from the supporting and obstructing data in (A), respectively. (C) Magnetic difference signal, calculated from the supporting and obstructing data in (A), respectively. (D) Sum of the supporting and obstructing signals in (C). The insets are beam camera images that show the pump-probe overlap for $l = 3$ (left) and $l = -3$ (right). The scale bars are 1 mm.
Fig. S6.

Experimental data of $|l| = 4$. For optimal visualization, the data points were connected with lines. The blue lines correspond to data from supporting experimental geometries while the red curves correspond to data from obstructing geometries. (A) Normalized and shifted (in time) experimental data from the four possible excitation geometries. (B) Nonmagnetic sum signal, calculated from the supporting and obstructing data in (A), respectively. (C) Magnetic difference signal, calculated from the supporting and obstructing data in (A), respectively. (D) Sum of the supporting and obstructing signals in (C). The inset is a beam camera image that shows the pump-probe overlap for $l = 4$ (the overlap was also checked for $l = -4$, however, the image was accidentally not saved). The scale bar is 1 mm.
Fig. S7.
Experimental data of $|l| = 5$. For optimal visualization, the data points were connected with lines. The blue lines correspond to data from supporting experimental geometries while the red curves correspond to data from obstructing geometries. (A) Normalized and shifted (in time) experimental data from the four possible excitation geometries. (B) Nonmagnetic sum signal, calculated from the supporting and obstructing data in (A), respectively. (C) Magnetic difference signal, calculated from the supporting and obstructing data in (A), respectively. (D) Sum of the supporting and obstructing signals in (C). The insets are beam camera images that show the pump-probe overlap for $l = 5$ (left) and $l = -5$ (right). The scale bars are 1 mm.
Experimental data of $|l| = 6$. For optimal visualization, the data points were connected with lines. The blue lines correspond to data from supporting experimental geometries while the red curves correspond to data from obstructing geometries. (A) Normalized and shifted (in time) experimental data from the four possible excitation geometries. (B) Nonmagnetic sum signal, calculated from the supporting and obstructing data in (A), respectively. (C) Magnetic difference signal, calculated from the supporting and obstructing data in (A), respectively. (D) Sum of the supporting and obstructing signals in (C). The insets are beam camera images that show the pump-probe overlap for $l = 6$ (left) and $l = -6$ (right). The scale bars are 1 mm.
Experimental data of $|l| = 7$ with a lower pump fluence than the data in Figs. S10, S11, and S12. For optimal visualization, the data points were connected with lines. The blue lines correspond to data from supporting experimental geometries while the red curves correspond to data from obstructing geometries. (A) Normalized and shifted (in time) experimental data from the four possible excitation geometries. (B) Nonmagnetic sum signal, calculated from the supporting and obstructing data in (A), respectively. (C) Magnetic difference signal, calculated from the supporting and obstructing data in (A), respectively. (D) Sum of the supporting and obstructing signals in (C). The insets are beam camera images that show the pump-probe overlap for $l = 7$ (left) and $l = -7$ (right). The scale bars are 1 mm.
Fig. S10.
Experimental data of $|l| = 7$ with a higher pump fluence than the data in Fig. S9 and a lower pump fluence than the data in Figs. S11 and S12. For optimal visualization, the data points were connected with lines. The blue lines correspond to data from supporting experimental geometries while the red curves correspond to data from obstructing geometries. (A) Normalized and shifted (in time) experimental data from the four possible excitation geometries. (B) Nonmagnetic sum signal, calculated from the supporting and obstructing data in (A), respectively. (C) Magnetic difference signal, calculated from the supporting and obstructing data in (A), respectively. (D) Sum of the supporting and obstructing signals in (C). The insets are beam camera images that show the pump-probe overlap for $l = 7$ (left) and $l = -7$ (right). The scale bars are 1 mm.
Fig. S11.
Experimental data of $|l| = 7$ with a higher pump fluence than the data in Figs. S9 and S10 and a lower pump fluence than the data in Fig. S12. For optimal visualization, the data points were connected with lines. The blue lines correspond to data from supporting experimental geometries while the red curves correspond to data from obstructing geometries. (A) Normalized and shifted (in time) experimental data from the four possible excitation geometries. (B) Nonmagnetic sum signal, calculated from the supporting and obstructing data in (A), respectively. (C) Magnetic difference signal, calculated from the supporting and obstructing data in (A), respectively. (D) Sum of the supporting and obstructing signals in (C). The insets are beam camera images that show the pump-probe overlap for $l = 7$ (left) and $l = -7$ (right). The scale bars are 1 mm.
Fig. S12.
Experimental data of $|l| = 7$ with a higher pump fluence than the data in Figs. S9, S10, and S11. For optimal visualization, the data points were connected with lines. The blue lines correspond to data from supporting experimental geometries while the red curves correspond to data from obstructing geometries. (A) Normalized and shifted (in time) experimental data from the four possible excitation geometries. (B) Nonmagnetic sum signal, calculated from the supporting and obstructing data in (A), respectively. (C) Magnetic difference signal, calculated from the supporting and obstructing data in (A), respectively. (D) Sum of the supporting and obstructing signals in (C). The insets are beam camera images that show the pump-probe overlap for $l = 7$ (left) and $l = -7$ (right). The scale bars are 1 mm.
Fig. S13.
Experimental data of $|l| = 8$ with a lower pump fluence than the data in Fig. S14. For optimal visualization, the data points were connected with lines. The blue lines correspond to data from supporting experimental geometries while the red curves correspond to data from obstructing geometries. (A) Normalized and shifted (in time) experimental data from the four possible excitation geometries. (B) Nonmagnetic sum signal, calculated from the supporting and obstructing data in (A), respectively. (C) Magnetic difference signal, calculated from the supporting and obstructing data in (A), respectively. (D) Sum of the supporting and obstructing signals in (C). The insets are beam camera images that show the pump-probe overlap for $l = 8$ (left) and $l = -8$ (right). The scale bars are 1 mm.
Fig. S14.
Experimental data of $|l| = 8$ with a higher pump fluence than the data in Fig. S13. For optimal visualization, the data points were connected with lines. The blue lines correspond to data from supporting experimental geometries while the red curves correspond to data from obstructing geometries. (A) Normalized and shifted (in time) experimental data from the four possible excitation geometries. (B) Nonmagnetic sum signal, calculated from the supporting and obstructing data in (A), respectively. (C) Magnetic difference signal, calculated from the supporting and obstructing data in (A), respectively. (D) Sum of the supporting and obstructing signals in (C). The insets are beam camera images that show the pump-probe overlap for $l = 8$ (left) and $l = -8$ (right). The scale bars are 1 mm.
Fig. S15.
Fitting of laser-induced demagnetization for different orders of photonic OAM that is not shown in the main manuscript. (A) $|l| = 2$, (B) $|l| = 3$, (C) $|l| = 4$, (D) $|l| = 6$, (E-G) $|l| = 7$ (different pump fluences), and (H-I) $|l| = 8$ (different pump fluences). The light blue and light red dots represent the experimental data for the supporting and the obstructing case, respectively. The black lines correspond to the common demagnetization behavior while the dark blue and red lines are the additional Gaussians that are added to the shared behavior to account for the supporting/obstructing influence of the optical OAM (with an offset of +1 for better visualization). The bright blue and red lines show the combinations of the shared behavior and the OAM influence.
Fig. S16.
Experimental data of $l = 0$ with different pump fluences. For optimal visualization, the data points in (A-C) were connected with lines. (A) Normalized and shifted (in time) experimental data from the two different directions of the external magnetic field. (B) Nonmagnetic sum signal, calculated from the data shown in (A). (C) Magnetic difference signal, calculated from the data shown in (A), respectively. (D) Fitting of the laser-induced demagnetization. The dots represent the experimental data while the lines correspond to the fits. The inset is a beam camera image that shows the pump-probe overlap. The scale bar is 1 mm.
Fig. S17.
The OAM order does not influence the demagnetization behavior that is shared by both OAM handednesses (black curves in Fig. 3 and Fig. S15 for \(|l| = 1 − 8\), curves in Fig. S16D for \(l = 0\)). The minimal magnetization (minimum of the black curves in Fig. 3 and Fig. S10 for \(|l| = 1 − 8\), minimum curves in Fig. S16D for \(l = 0\), this is not \(M_{\text{min}}\) from equation (S1)) is plotted vs. the delay time \(t_{\text{min}}\) at which this minimum is reached. All OAM orders show a similar behavior; the value of \(t_{\text{min}}\) increases for a decreasing minimal magnetization (meaning a stronger quenching). This supports the assumption of our model, that the OAM of light adds an additional contribution to the ultrafast demagnetization that can be well approximated with a Gaussian. Because the minimal magnetization value is not a fitting parameter, no error limits are given. The errors of \(t_{\text{min}}\) are the fitting errors of \(t_0\). For \(|l| = 6\), no data point is plotted because the minimal magnetization was not reached in the measured delay time range.
Table S1.
Overview of the parameters and corresponding errors that are visualized in Fig. 4 and Fig. S17. The values for both $t_{min}$ and the center of the OAM contribution ($= t_{center} - t_0$) are given with respect to $t_0$. The minimal magnetization as well as $t_{min}$ are not fitting parameters but were extracted directly from the fitting curves, thus there is no error given for the minimal magnetization while $\Delta t_{min} = \Delta t_0$. The error of the center of the OAM contribution with respect to $t_0$ was calculated via $\Delta(t_{center} \text{ of OAM contribution}) = \Delta t_{center} - \Delta t_0$.

| OAM order $|l|$ | Corresponding figures | $t_0$ (fs) | Minimal magnetization (norm.) | $t_{min}$ (fs) | $\Delta t_{min}$ (fs) | Center of OAM contribution (fs) | $\Delta$(Center of OAM contribution) (fs) |
|---|---|---|---|---|---|---|---|
| 0 | S16 | 0.4 | 0.82 | 200.6 | 29.7 | − | − |
| 0 | S16 | 0.0 | 0.75 | 210.0 | 22.9 | − | − |
| 0 | S16 | 0.3 | 0.69 | 307.7 | 18.2 | − | − |
| 0 | 2, S16 | -0.2 | 0.58 | 336.2 | 10.9 | − | − |
| 0 | S16 | 0.2 | 0.49 | 389.8 | 8.7 | − | − |
| 1 | 3, S2 | 0.3 | 0.65 | 277.7 | 10.4 | 213.9 | 22.5 |
| 2 | S3, S15A | 0.3 | 0.40 | 376.7 | 5.0 | 162.8 | 11.4 |
| 3 | 3B, S4 | 0.3 | 0.71 | 301.7 | 6.1 | 135.9 | 18.6 |
| 3 | S5, S15B | 0.5 | 0.53 | 282.5 | 6.7 | 103.0 | 17.2 |
| 4 | S6, S15C | -0.1 | 0.55 | 388.1 | 8.1 | 126.8 | 22.6 |
| 5 | 2A, 3C, S7 | 0.1 | 0.58 | 338.9 | 7.4 | 121.5 | 14.6 |
| 6 | S8, S15D | -0.5 | 0.35 | − | 19.6 | 100.9 | 24.5 |
| 7 | 2B, 3D, S9 | 0.4 | 0.59 | 339.6 | 8.6 | 141.5 | 14.7 |
| 7 | S10, S15E | 0.2 | 0.56 | 295.8 | 8.8 | 158.9 | 13.8 |
| 7 | S11, S15F | 0.2 | 0.54 | 405.8 | 7.9 | 113.2 | 14.7 |
| 7 | S12, S15G | 0.4 | 0.49 | 380.6 | 9.2 | 97.8 | 15.1 |
| 8 | S13, S15H | -0.9 | 0.70 | 208.9 | 23.1 | 81.0 | 43.7 |
| 8 | S14, S15I | 0.2 | 0.44 | 416.8 | 5.0 | 61.7 | 23.6 |