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ABSTRACT

We investigate the impact of non-collinear dual optical excitation on magnetization precession in a permalloy thin film using two ultrashort laser pulses. By analyzing the magnetization dynamics using time-resolved magneto-optical methods, we find that excitation with two ultrashort optical pulses introduces a long-lasting modification of the electron system, as indicated by a sizable decrease in the precession frequency and a significant increase (approximately 25%) in the decay time. Our results reveal that the observed effect strongly depends on the respective polarizations of the two excitation pulses and the time delay between the two optical pulses. Our findings indicate the occurrence of a nonlinear opto-spin effect during photoexcitation with two interfering optical pulses, which can potentially be observed in various materials and at different photon wavelengths.

I. INTRODUCTION

The interaction of light with matter is one of the most fundamental processes in nature. From the absorption and emission of light by atoms and molecules to the propagation of light through materials, the properties of matter are intimately tied to the behavior of light and vice versa. Researchers have made significant progress in understanding light–matter interaction, particularly with the development of ultrafast lasers that generate light pulses with a duration on the order of femtoseconds, which makes it possible to study the dynamics of the interaction on a time scale that matches the response of the material.[1] However, one of the key challenges in studying and controlling light–matter interaction is the response of the material occurring on short time scales (femtoseconds to picoseconds).

Femtosecond laser pulses have become an increasingly important tool for controlling material properties on ultrashort time scales. All-optical magnetization reversal,[2–4] evidence of light-induced superconductivity,[5,6] and optical-driven phase transitions[7,8] are only some examples. While ultrashort pulses have been tuned over a wide range of wavelengths, allowing selective excitation of the electron and lattice degrees of freedom, a vast majority of studies use a single laser pulse to pump the system. Very few studies have explored the possibility of manipulating matter with two or more ultrashort pulses.[9–14] In such a two-step excitation process, the first pump pulse promotes the material to an intermediate excited state, while the second pump pulse further drives the system out of equilibrium. In certain cases, this approach allows for more efficient control of the material properties.

It is generally believed that excitation with two optical pulses that overlap in time and space at the microscopic level behaves like single-pulse excitation. In other words, the response of an electron when excited with two non-collinear propagating photons is expected to be the same as when it is excited with photons that propagate collinearly. Nonlinear optical effects such as the formation of transient gratings,[15–17] difference, or sum frequency generation could take place, but the long-lasting effect of excitation on the material is expected to follow simply the dependence determined

by the strength of the electric or magnetic field of the incoming radiation.

Here, we demonstrate that optical excitation with two non-collinear light pulses can cause an unexpected response in a magnetic material. We show that the amplitude, decay time, and frequency of magnetization precession in magnetically ordered alloys change when the dynamics is triggered by two non-collinear interfering optical pulses. These observations cannot be explained by the occurrence of a transient grating and indicate the existence of an opto-spin effect that results in modified intrinsic magnetic properties of the material persisting much longer than the duration of the two coherent optical pulses.

We studied the effect of dual non-collinear optical excitation on the widely explored physical process of laser-induced magnetization precession.[18,19] This process involves the interaction between light, electrons, and eventually spins, making the magnetization precession an ideal test bed for our considerations. The mechanism for triggering magnetization oscillations with an ultrashort laser pulse is based on the heat-induced reduction of magnetization M,[20] resulting in a local change in the magnetic anisotropy and subsequent pointing of the magnetization away from the equilibrium position. This launches a magnetization precession that can be described by the Landau–Lifshitz–Gilbert equation and the excitation of spin waves.[21] We have extended this simple approach by implementing a second excitation pulse and varying the delay between the two optical pump pulses.

II. EXPERIMENTAL DETAILS

The material studied in our experiments was a permalloy (Py) thin film deposited on top of a SiO_2 substrate. Py, a $Ni_{80}Fe_{20}$ alloy with a thickness of approximately 20 nm, was prepared using a thermal evaporation method at a base pressure of 1×10^{-6} mbar and was capped with a 2-nm-thick layer of gold to prevent oxidation. Py has attracted considerable interest due to the magnetic monopole-like behavior observed in nanoscale patterned Py structures (e.g., Py artificial spin ice),[22–24] the optical excitation of propagating spin waves,[25,26] and the use of Py as a working material for spintronic applications.[27] These observations benefited from the very low magnetic damping in Py.[28,29]

The experimental configuration is shown in Fig. 1(a). Ultrashort light pulses were delivered from a Yb-fiber laser operating at a fundamental wavelength of $\lambda = 1030$ nm and at a repetition rate of 50 kHz. Optical excitation was performed with two linearly polarized 300 fs pulses with a wavelength of $\lambda = 1030$ nm and a spot size of 250 μm, with incidence angles of 2° and 60° for pump 1 and pump 2, respectively. Linearly polarized optical probe pulses with a wavelength of $\lambda = 515$ nm and a spot size of 100 μm were obtained by frequency doubling of the fundamental wavelength of the laser. The probe beam was at an angle of incidence of approximately 5° to the surface normal. Pump–probe delay, used later, refers to delay between pump 1 and probe beams. The magnetization dynamics was studied by analyzing the Kerr rotation of the polarization plane of the reflected probe beam using a balanced detection scheme. In this geometry, most of the magnetic signal comes from the polar magneto-optical Kerr effect (MOKE), which is sensitive to the out-of-plane component of magnetization M_Z. An external magnetic field of $H = 1$ kOe was applied parallel to the sample surface.

III. RESULTS

An example of the magnetization dynamics excited only with pump 1 and pump 2 is given in Fig. 1(b), which exhibits the oscillatory behavior of the M_Z component caused by magnetization precession. We detect precession dynamics up to 920 ps after excitation, which is the longest available delay range in our experimental setup. Considering the different absorption for different incidence angles and, hence, different incident fluences of $F = 0.72$ mJ/cm^2 for pump 1 and $F = 1.3$ mJ/cm^2 for pump 2, there is no difference in the dynamics excited by P-polarized or S-polarized pump 1 or P-polarized pump 2, as expected for a thermal excitation mechanism. The time-resolved traces were fit with a damped sinusoidal function,

$$\Delta M(t) = Ae^{-\frac{t}{\delta}} sin(2\pi(f + \Delta ft)t - \varphi), \quad (1)$$

where A is the amplitude of oscillations, f is the frequency, Δf is the coefficient to cover the temporal change of the precession frequency upon recovering magnetization, and δ is the decay time. The amplitude and frequency of the magnetic oscillations change monotonically with increasing fluence [see Figs. 1(c) and 1(d)], while the decay time remains constant [Fig. 1(e)]. It is important to note that magnetization precessions excited with P- or S-polarized pump 1 are nearly identical [see Figs. 1(c)–1(e)], indicating that the absorption of pulses from pump 1 at nearly normal incidence is the same when varying the polarization.

The excitation of the sample with two optical pulses of the same fluence, $F = 1.5$ mJ/cm^2, leads to different magnetization dynamics. While the dynamics closely resemble those induced by a single optical pulse when pump 1 is S-polarized and pump 2 is P-polarized, a notable divergence occurs when both pump pulses are P-polarized, resulting in distinct oscillations. The frequency, amplitude, and decay time of these oscillations [cf. Eq. (1)] as a function of the delay time between the two pump pulses are shown in Figs. 2(b), 2(d), and 2(e), respectively. Each panel delineates the dependencies for both the P–P configuration (where both pump pulses have P polarization) and the S–P configuration (where pump 1 is S-polarized and pump 2 is P-polarized). Remarkably, all three parameters change when the two pump beams temporally overlap in the P–P configuration, while no changes are observed when the polarizations are orthogonal. We note that the total absorption is the same for both the P–P and S–P polarization configurations because we only change the polarization of pump 1, which has no polarization dependence on the absorption due to the nearly normal incidence. The decay time of precession, when excited with P-polarized pump beams, exhibits an approximately 25% increase compared to that of the configuration with orthogonal linear polarization, and the frequency decreases by approximately 1%. While a change in oscillation frequency could be anticipated due to the different degrees of demagnetization [see Fig. 1(c)], the observed frequency change surpasses expectations based solely on the observed increase in the amplitude of magnetization precession in the P–P configuration in Fig. 2(d). Consequently, we attribute this observed frequency change to an

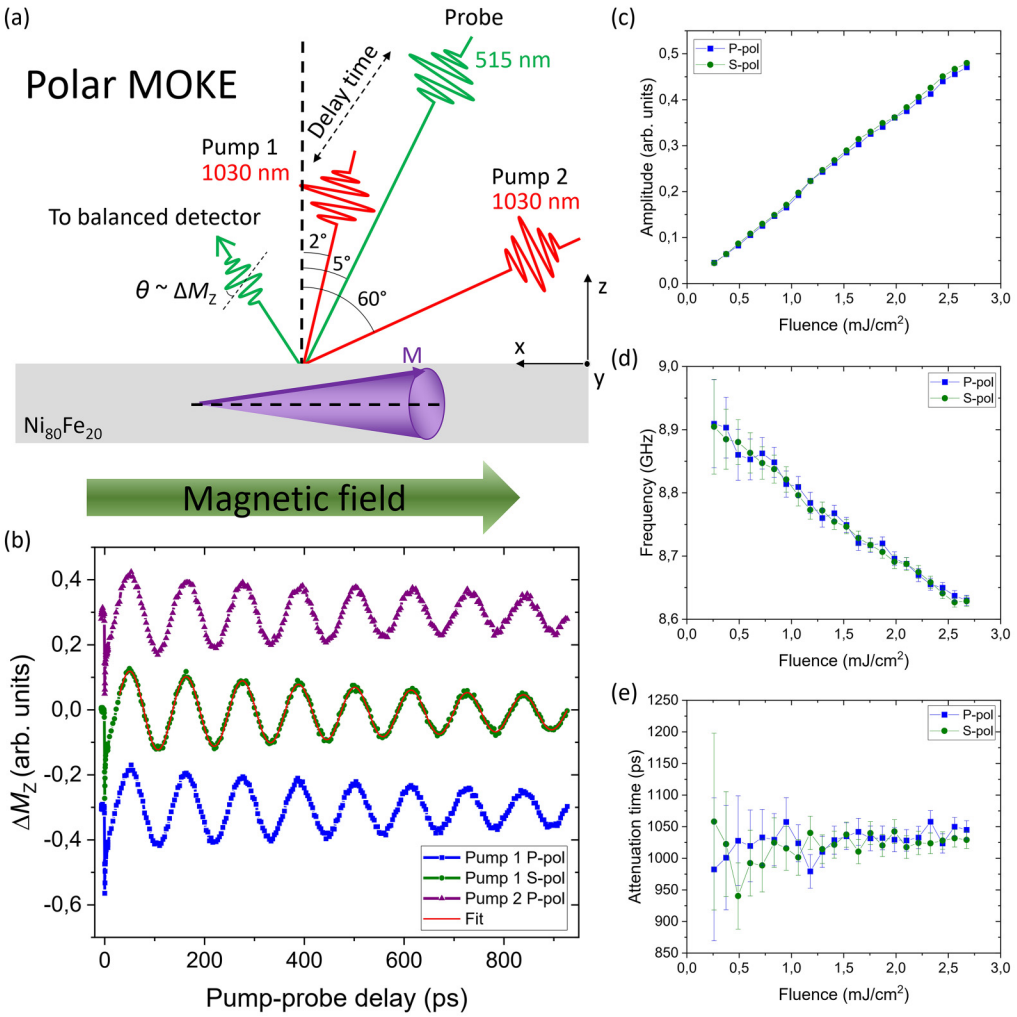

FIG. 1. (a) Schematic illustration of the experimental geometry. The permalloy sample is excited by two non-collinear laser pumps, and the magnetization dynamics is probed with the polar magneto-optical Kerr effect (P-MOKE). (b) Time-resolved magnetization dynamics after single-pulse excitation for different pump polarizations with an incident fluence of F = 0.72 mJ/cm^2 for pump 1 and F = 1.3 mJ/cm^2 for pump 2. While the oscillation curves are the same in frequency and amplitude, indicating the equivalence of optical excitation with both pumps, a higher fluence for pump 2 is required due to different absorption at a 60° incidence angle. The curves are vertically shifted for better visibility. The red solid line is fit to Eq. (1). (c) Amplitude, (d) frequency, and (e) decay time of magnetization precession as a function of pump 1 fluence for different polarizations.

effect associated with non-collinear dual optical excitation, which is specifically linked to interference between the two pump beams. In Figs. 2(c) and 2(f), we present panels illustrating the frequency and decay time as a function of the polarization direction of pump 1 for $\Delta\tau = 0$ ps. Both dependencies follow the superposition of the E-field vector projections of the two pumps. Furthermore, looking at magnetization precession in the S–P configuration, we can say that the excitation strength with two pump beams roughly corresponds to a single pulse excitation with a fluence of approximately $F = 2.5$ mJ/cm^2. Thus, the possible effect on magnetization dynamics from non-linear absorption effects in this fluence range should be also seen in the single pump excitation regime. Indeed, the frequency and decay time after single-pulse excitation with $F = 2.5$ mJ/cm^2 are nearly the same as those after dual-pump excitation in the S–P configuration, while the dynamics in the P–P configuration is different.

Two non-collinear optical pump beams that overlap in time can induce transient grating through interference between the two pulses.[15–17] The spatial modulation of the excitation intensity in a transient grating can potentially alter the behavior of magnetization precession. To evaluate the impact of a transient grating, we varied the angle θ between the pump pulses, thereby altering the period of the transient grating. The calculated spatial distribution of the electric field square with the finite-difference time-domain method

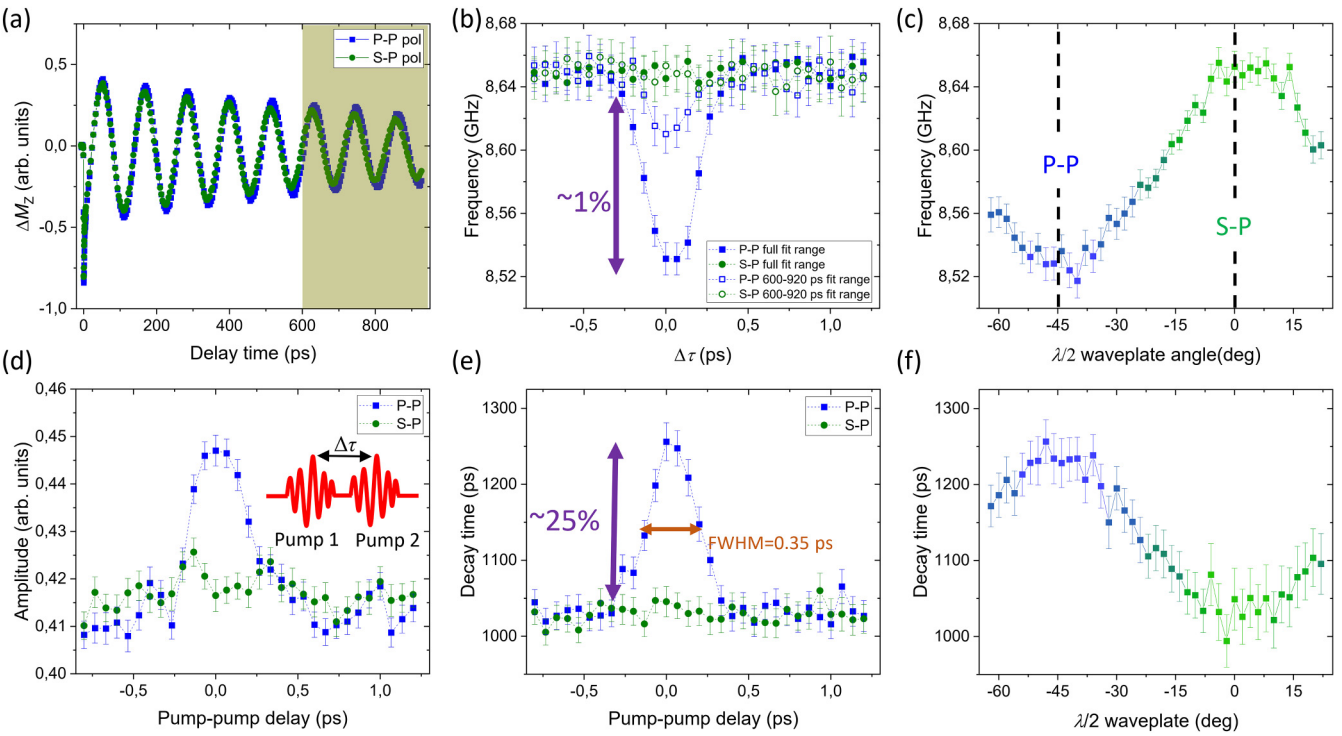

FIG. 2. (a) Time-resolved magnetization dynamics after dual-pulse excitation with different pump 1 polarizations and with fluencies F = 1.3 mJ/cm^2 for both pump beams. (b), (d), and (e) Frequency, amplitude, and decay time of magnetization precession mode, respectively, as a function of the delay Δτ between two pump pulses with different polarization configurations. (c) Frequency and (f) decay time of the magnetization precession as a function of the pump 1 polarization direction, determined by the angle of the half-wave plate.

using parameters similar to those described in the experimental conditions and for different separation angles is depicted in Fig. 3(a). With $\theta = 28°$, a well-defined interference pattern with clear maxima and minima and a period $d = 1.92\,\mu$m is formed. In contrast, for $\theta = 58°$, the interference pattern is less pronounced, with $d = 1.15\,\mu$m close to the wavelength of the pump beams, suggesting a significantly smaller effect of the transient grating for $\theta = 58°$.

The experimental data for the magnetization dynamics for $\theta = 28°$ configuration are presented in the supplementary material, and those for $\theta = 58°$ are given in the main text above. When comparing two θ configurations is important to consider changes in both frequency and decay time as both changes in P–P geometry. Thus, we discuss the Gilbert damping parameter $\alpha = 1/(2\pi f\delta)$, as it contains both the frequency and decay time when comparing two θ geometries. The data from Figs. S3(c), S3(d), S2(b), and S2(e) in the supplementary material were used to calculate α as a function of $\Delta\tau$ for different θ, as shown in Figs. 3(b) and 3(c). The different value of parameter α for two θ geometries when two pump pulses are separated in time and in S–P geometry is discussed in supplementary material. For both θ configurations, there is a clear but opposite change in α for P–P pump polarizations when the beams overlap in time. In both cases, the change in α is primarily caused by alterations in the decay time [see Figs. S3(c) and S2(e) in the supplementary material], despite the concurrent decrease in the frequency of approximately 1% observed for both θ configurations [see Figs. S3(d) and S2(b) in the supplementary material]. The spatial modulation of excitation leads to magnetic oscillations with slightly different frequencies of interference maxima and minima, given that the precession frequency is a function of pump power [see Fig. 1(d)]. The dephasing between oscillations at maxima and minima during recovery likely contributes to the suppression of magnetization precession for $\theta = 28°$. Following these arguments, for $\theta = 58°$ configuration, one would expect a smaller increase in α, as the difference between the maxima and minima is much smaller. Surprisingly, a decrease in α is observed for $\theta = 58°$, suggesting that the observed increase in decay time and decrease in the frequency of magnetization precession result from interfering optical pulses but not from the transient grating. Furthermore, when performing the fitting procedure in the delay range of 600–920 ps [refer to the shaded area in Fig. 2(a)], where the transient grating should have already dissipated,[30] a change in oscillation frequency is still detected [Fig. 2(b) open circles]. Finally, the periodic excitation pattern might promote the excitation of additional spin precession modes that will interfere with the main one. Such a coupling could be seen as alternated magnetization precession. However, the increase in decay time in such a situation is highly unlikely and the absence of additional modes is confirmed by fast Fourier transformation analysis of the time traces from Fig. 2(a) [see Fig. 3(d)].

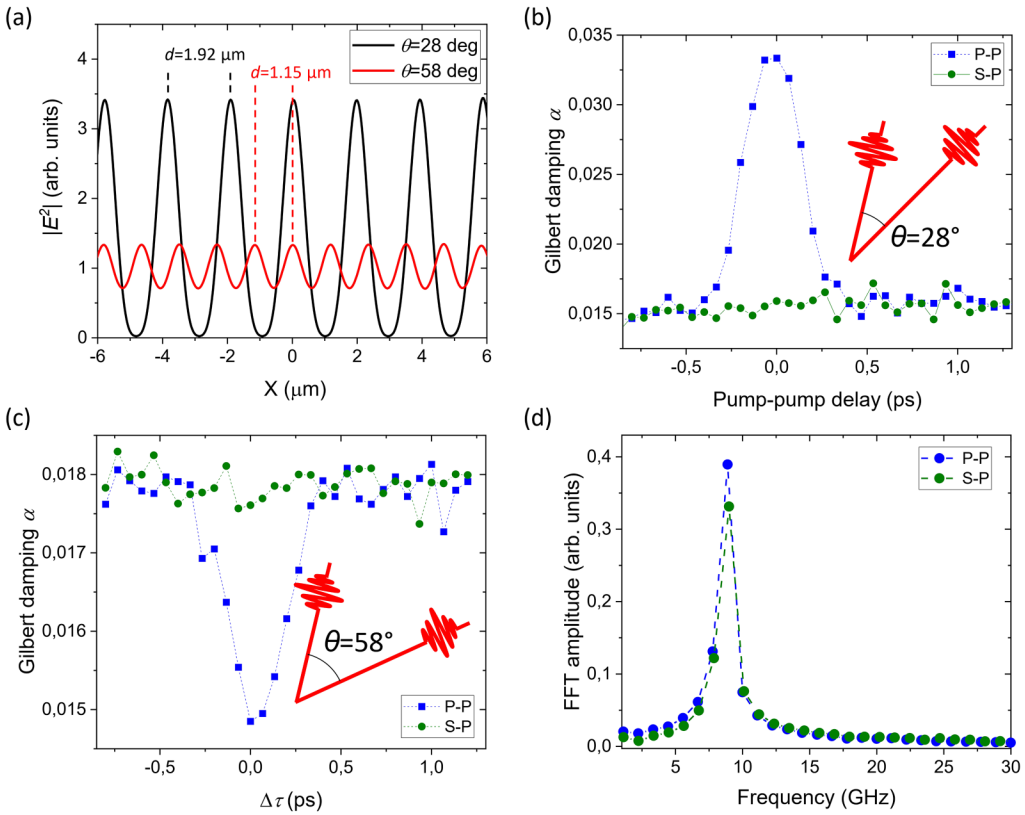

FIG. 3. (a) Calculated spatial distribution of the electric field square $|E^2|$ when two optical pulses impinge the sample with different separation angles θ. (b) and (c) Gilbert damping parameter α as a function of delay between two pump pulses Δτ for configurations with θ = 28° and θ = 58°, respectively. (d) Fast Fourier transformation spectra of time traces from Fig. 2(a).

IV. DISCUSSION

The observed changes in frequency and decay time suggest that optical excitation with two pump pulses changes the magnetic configuration of the material. We attempt to explain the unusual coupling of the dual pump pulses to the magnetization dynamics considering a recently derived relativistic light–spin interaction term in the Hamiltonian,[31]

$$\mathcal{H} = \frac{e^2\hbar}{8m^2c^2\omega}\boldsymbol{\sigma}\cdot \mathrm{Re}[-i(\boldsymbol{E}\times\boldsymbol{E}^*)] = -g\mu_B\boldsymbol{\sigma}\cdot\boldsymbol{B}_{opt}\,, \tag{2}$$

where $\boldsymbol{\sigma}$ denotes the Pauli spin matrices, $\boldsymbol{E}$ is the total electric field, and $\boldsymbol{B}_{opt}$ is the optomagnetic field caused by the electric fields with frequency ω. Note that the form of this interaction term is similar to that of the inverse Faraday effect,[32] wherein, however, a single circularly polarized pulse is commonly employed. Here, the total electric field results from the two pump pulses, i.e., $\boldsymbol{E} = \boldsymbol{E}_{pu1} + \boldsymbol{E}_{pu2}$. For the pump pulses, we assume plane waves modulated with a Gaussian envelope. This gives for the optomagnetic field (see supplementary material),

$$\boldsymbol{B}_{opt} = \frac{e^2\hbar}{8m^2c^2\omega g\mu_B}\exp\left[-\frac{t^2}{2\Gamma^2}-\frac{(t-\Delta\tau)^2}{2\gamma^2}\right]\mathrm{Re}[i(\boldsymbol{E}^0_{pu1}\times\boldsymbol{E}^{0*}_{pu2}e^{i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega\Delta\tau)} - \boldsymbol{E}^{0*}_{pu1}\times\boldsymbol{E}^0_{pu2}e^{-i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega\Delta\tau)})]. \tag{3}$$

The width of the pulses is determined by γ and Γ, $\boldsymbol{k} = \boldsymbol{k}_{pu1} - \boldsymbol{k}_{pu2}$ is the resultant wave vector, which is nonzero for noncollinear beams, and $\boldsymbol{E}^0_{pu1}$, $\boldsymbol{E}^0_{pu2}$ are the electric field vectors of the pulses. It can be recognized from Eq. (3) that the optomagnetic field is maximal when the two pulses fully overlap, i.e., $\Delta\tau = 0$, which is consistent with the measurements shown above.

Next, we analyze the dependence of the optomagnetic field on pump polarization. Starting with the P–P configuration, we compute a nonzero $\boldsymbol{B}_{opt}$ aligned predominantly along the y direction (see supplementary materials for details). The non-collinearity of the beams is important for obtaining a nonzero $\boldsymbol{B}_{opt}$. The torque due to the optomagnetic field, $\boldsymbol{T} = \boldsymbol{M} \times \boldsymbol{B}_{opt}$, is dominantly in the out-of-plane z direction. An increase in the amplitude of the magnetization precession could be caused by the additional torque in the z direction. The S–P configuration leads to a nonzero $\boldsymbol{B}_{opt}$ along the z direction and a torque $\boldsymbol{T}$ along the in-plane y direction. This direction, however, cannot lead to a polar MOKE signal, as this signal is sensitive only to out-of-plane magnetization excursions. The observed dependencies on pump polarizations are, thus, consistent with those expected from the light–spin interaction (2). The light-induced torque changes the phase of magnetization precession. Figure 4 shows the phase of magnetization precession mode in P–P and S–P configurations as a function of pump–pump delay. There is a clear phase shift of magnetization precession for $\Delta\tau = 0$ in the P–P configuration that confirms the above statements.

The light–spin interaction (2) can be seen as an alteration of the spin angular momentum of electrons with the angular momentum of photons. This momentum alteration would change the magnetic couplings and effectively the magnetic moment of the system, leading to an alteration of the magnetic oscillation mode. However, the long-liveness of the induced effect is surprising. A recent investigation using single circularly polarized laser pulses revealed dynamics in Co/Pt layers induced by the photon spin angular momentum lasting for more than 60 ps but did not analyze modifications of Gilbert damping.[33] Here, our analysis shows that the dynamics in a Py film are almost 1 ns long, which indicates that the induced effect is long-lived compared to the 0.35 ps $\Delta\tau$ window where the two pump pulses overlap, which is the acting time of the photomagnetic field $\boldsymbol{B}_{opt}$. The light-induced angular momentum change is expected to be transferred from electrons to the lattice, but even though energy is transferred between electron and phonon subsystems on a picosecond time scale,[1] this does not imply that angular momentum is transmitted equally fast. Additionally, even when the angular momentum is passed on to the lattice, it is still in the system, and it can affect the magnetization dynamics. Finally, we clarify that data in Fig. 4 and described above Hamiltonian do indicate that two linearly polarized non-collinear pulses have similar action to the electron angular momentum as a single circularly polarized light pulse, but it does not explain how the angular momentum of the system could remain modified for such a long time.

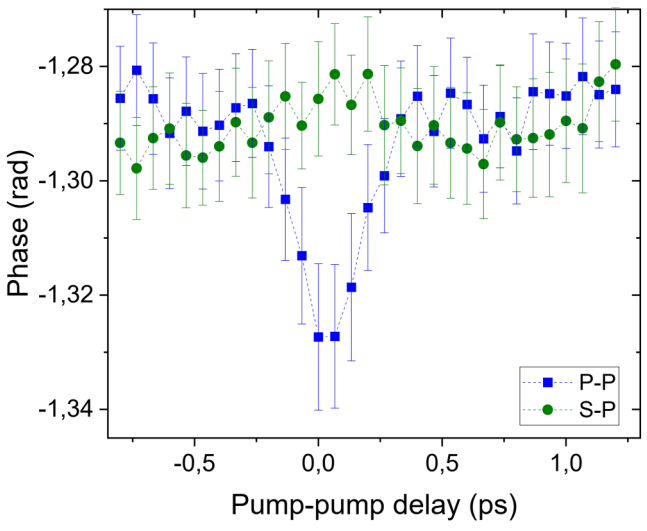

FIG. 4. Phase of magnetization precession mode as a function of delay between two pump pulses for configuration with θ = 58°.

Several prior theoretical investigations have aimed to reveal the interaction of light with spins on the time scale of optical excitation, including predictions of the coupling of the angular moment of light to the spin.[34,35] A similar coupling as in Eq. (2) between the electromagnetic angular moment density and magnetic moments in a solid was phenomenologically proposed.[36] There is experimental evidence of relativistic spin–photon coupling reported in Ref. 34, which was later supported by theoretical investigations,[35,37] albeit on the basis of different relativistic interactions. The study of Bigot *et al.*,[34] thus, provided evidence of an optomagnetic field acting on the electron during pump excitation. However, a single laser pump was used, and no long-lasting effects were reported.

V. CONCLUSION

In conclusion, we investigated ultrafast magnetization precession in a permalloy thin film using two ultrashort excitation pulses. Our study has demonstrated that using two non-collinearly propagating optical pulses that interfere can elicit long-lasting changes in the electronic properties of the material, seen as alterations in the decay time and frequency of magnetization precession. The observed effect is notably dependent on the relative polarization of the two pulses and is maximized when the excitation pulses overlap in time. The results presented here challenge our current understanding of light–matter interactions. Theoretically, we explore the concept of relativistic light–spin interaction, conceptualized as an optomagnetic field, acting on the time scale of the pump beam duration. The long-lasting modification of the decay time and frequency on a nanosecond time scale is surprising in view of the common understanding of light-induced magnetic processes where relaxation of laser-imparted energy should occur on a ps time scale. Hence, we hypothesize that the observed deviation in magnetization dynamics might arise from the fact that the light-induced modification of the angular momentum remains longer in the material, in either the electronic or the phononic subsystems, and can modify the magnetization dynamics. The key feature to achieve such modification is the optical excitation of the medium with interfering light. This work not only introduces new possibilities for the control and manipulation of magnetic states in materials but also suggests potential applications for manipulating electron spins in unforeseen ways. Additionally, our findings offer insights into fundamental interactions between light and matter, with

implications that may extend beyond the realm of magnetism. In particular, the significance of considering optical excitation with interfering light for studying and controlling material properties is underscored by our results. Our results encourage further investigations to unravel the underlying mechanisms of this effect and explore its potential applications across diverse material systems.

SUPPLEMENTARY MATERIALS

See the supplementary material for additional experimental data on dual pump excitation of magnetization precession with different angles between two pump beams and a detailed theoretical derivation of the opto-magnetic field resulting from optical excitation with ultrashort laser pulses.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Sergii Parchenko: Conceptualization (lead); Data curation (lead); Formal analysis (lead); Investigation (lead); Writing – original draft (lead); Writing – review & editing (lead). **Davide Pecchio:** Investigation (equal); Writing – review & editing (equal). **Ritwik Mondal:** Investigation (equal); Writing – review & editing (equal). **Peter M. Oppeneer:** Investigation (equal); Writing – review & editing (equal). **Andreas Scherz:** Investigation (equal); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

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

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