Interplay of heating and helicity in all-optical magnetization switching

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Contrasting hypotheses have been made about the role played by laser heating and photon helicity in all-optical switching. Here we present an experiment that distinguishes between heating- and helicity-driven effects. We show that even though a minimum amount of circularity is needed to switch, heating contributes to the process. Moreover, we show that the helicity information carried by the exciting laser pulses is more easily transferred into the magnetic material at lower temperatures and that it persists in GdFeCo for at least some picoseconds after optical excitation.

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The interaction between light and magnetic materials is known to cause a variety of different effects. One of the most famous is the magneto-optical Faraday effect, which results in a change of the light polarization after transmission through a magnetic material. The inverse effect is also possible, meaning that the polarization of the light affects the magnetic state of the sample.[1–3] For ultrashort laser pulses this was proven in 2005 by Kimel *et al*., who showed that the helicity of the exciting laser pulse influences the magnetic material response.[4] Furthermore the authors of Ref. 4 were able to deterministically switch the magnetization of a ferrimagnetic GdFeCo sample by using a single 40 fs circularly polarized laser pulse without any additional external magnetic field.[5] This effect is known and referred to here as "all-optical" or "optomagnetic" switching.

The interaction of ultrashort laser pulses with magnetic materials has been classified into three main categories,[6] according to the underlying microscopic mechanism: (1) thermal effects, (2) nonthermal photomagnetic effects, and (3) nonthermal optomagnetic effects. Thermal effects (1) are induced by the heating of the electrons, spins, and lattice systems. They include, for example, the ultrafast demagnetization phenomenon[7–15] as well as heat-assisted magnetic recording.[16–22] Recently even complete magnetization switching due to ultrafast heating by linearly polarized laser pulses and without an applied external magnetic field was demonstrated.[23] In contrast, the proposed category of nonthermal photomagnetic effects (2), which includes, for example, the photomagnetic modification of magnetocrystalline anisotropy,[24] does not rely on heating but requires the absorption of photons. Finally, nonthermal optomagnetic effects (3) are based neither on heating nor on photon absorption. An example is the optical excitation of magnetic precession,[4] which can be explained phenomenologically by inverse optomagnetic effects, or on a more microscopic basis in terms of a spin-flip stimulated Raman process.[25]

Whether all-optical switching can be assigned to one of the introduced categories (1)–(3) is still a matter of debate, since its microscopic origin is a very controversial issue.[26–29] In particular, the role of heating within the switching process is still unclear. For example, Stanciu *et al*. proposed that the switching process is a combination of two effects, namely, heating of the spin system close to the Curie temperature (which is connected to the heating of the lattice) and action of the circular light field as a magnetic field pulse.[5] Accordingly, all-optical switching should be sorted into categories (1) and (3). In contrast Hohlfeld *et al*. suggested that the light helicity alone can drive the switching process,[30,31] thus assigning it to category (2). In both cases, it is still unclear where the helicity information carried by the light should be stored in the material during the typical switching times of some 10 ps.[26] Identifying such an "helicity storage" is a nontrivial question, especially if one considers that the photoexcited electron-hole pairs are known to dephase within around 2 fs,[32] whereby afterward any phase information of the exciting laser is lost.

Here we present an experimental approach allowing us to separate heating- and helicity-driven effects within the switching process. We performed repetition-rate-dependent measurements of the minimum threshold fluence needed to obtain all-optical switching. Such investigations are extended by introducing a so-called σ-π *experiment*, where the pulse inducing all-optical switching is split in one circularly (σ) and one linearly (π) polarized pulse (see Fig. 1). The σ pulse is used to carry the helicity information, while the π pulse heats the sample. Performing this division, we are able to demonstrate that moderate heating contributes to the switching process, while concomitantly a minimum amount of circularity is needed. We find that the helicity transfer to the material is more efficient for lower phonon temperatures and that for GdFeCo the decay time of the material-specific helicity storage must be at least on the order of some picoseconds.

The experiments were carried out on a multilayer amorphous ferrimagnetic rare earth-transition metal (RE-TM) sample with out-of-plane magnetization, $Gd_{24}Fe_{66.5}Co_{9.5}$. It was grown by magnetron sputtering with the detailed structure glass/AlTi (10 nm)/SiN (5 nm)/RE-TM(20 nm)/SiN (60 nm). The compensation temperature is $T_{comp} = 280$ K, and the Curie temperature is around 500 K.

To detect the magnetic domains we used a Faraday imaging setup consisting of a white-light source, a crossed polarizer pair, and a CCD camera.[27] To switch the magnetization within a part of a homogeneously magnetized domain we utilized laser pulses with the following characteristics: For the repetition rate (ν_r)-dependent measurements we utilized a ps laser at a central wavelength of 532 nm with a pulse duration between 9 and 13 ps, slightly changing with ν_r, which could be varied from 1 to 500 kHz. The pulses were circularly polarized by a zero-order quarter-wave plate. For the σ-π experiments

we used a fs-amplifier system running at 5.2 kHz, respectively 6.5 kHz, at a central wavelength of 780 nm. The pulse duration could be changed from ≈90 fs to 2 ps at the sample position by adjusting the compressor in the amplifier. The beam was split with a 50/50 beam splitter to obtain the σ and π beam paths. The intensity of each beam could be separately adjusted by a gray filter wheel and the combination of a half-wave plate and a polarizer. Additionally, the σ path was circularly polarized by a zero-order quarter-wave plate. By means of a delayline the σ and π pulses were temporally delayed with respect to each other with a variable delay up to 330 ps. Both pulses were focused and spatially overlapped at the sample position. While the σ pulse impinged on the sample at normal incidence (0°) there was a small angle of less than 10° for the π pulse.

All-optical, meaning helicity-dependent, switching is known to work only in a relatively narrow fluence window.[26] Accordingly, two threshold fluences exist: The lower threshold fluence describes the transition from "no switching" to "all-optical switching," while the upper threshold fluence separates the "all-optical switching" range from the "pure thermal demagnetization" range. For the latter case the magnetization is affected independently of helicity, resulting in a multidomain structure, which is attributed to sample heating above the Curie temperature. In our experiments the physical observable is the minimum threshold fluence F_{min} that is needed to switch all-optically.[33] The laser beam was swept over a (homogeneously magnetized) area of the sample (by moving the sample) to minimize the influence of spatially different sample properties, and the switching behavior was double-checked by varying the combinations of helicity and sample magnetization (for a more detailed description of the measuring procedure see Ref. 27).

Figure 2 shows the dependence of F_{min} on the repetition rate of the exciting circularly polarized laser pulses, which was changed from single shot ($\nu_r = 0$) to 500 kHz.[34] F_{min} clearly decreases with increasing ν_r. Considering that higher repetition rates cause a stronger accumulative heating of the sample (electronic *and* phononic system), this result indicates that the switching process is favored by heating.

To investigate the interplay of heating and helicity in more detail, we separated the effects of heat and helicity on the switching process by implementing the σ-π experiments as described in the following. First, the minimum threshold fluence F_{min} required to switch all-optically with only one σ pulse (called "one pulse switching"; see Fig. 1) was determined. Then the fluence of this σ pulse (F_σ) was decreased until switching could not be achieved anymore. At this point, a π pulse was added with temporal and spatial overlap. By carefully adjusting the fluence of the π pulse (F_π), it was then possible to switch the magnetization again (called "two-pulse switching"; see Fig. 1). Thus, it is possible to switch all-optically for $F_\sigma < F_{\mathrm{min}}$ if in addition a heating π pulse is spatially and temporally overlapped to the σ pulse. This means that the threshold for one-pulse switching is not only determined by the number of circularly polarized photons, but also by a certain amount of heating. In other words, the pulses used for one-pulse (σ) switching have more circularity than intrinsically needed. By further increasing F_π above a certain threshold, pure thermal demagnetization (helicity independent) was observed. This indicates that strong heating

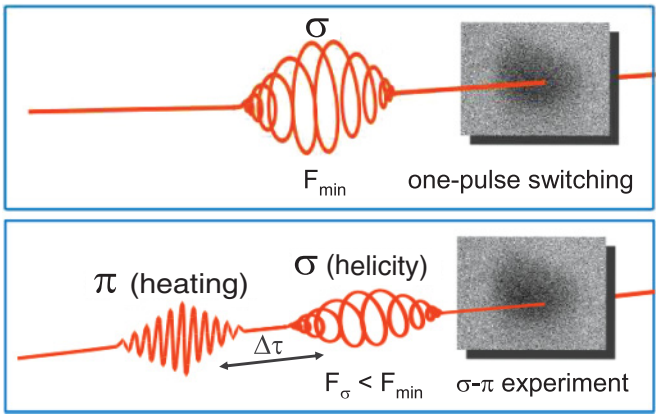

FIG. 1. (Color online) Scheme of the σ-π experiment (two-pulse switching) compared to one-pulse switching. A linearly and a circularly polarized pulse are temporally delayed and spatially overlapped at the sample position. The switching behavior for the combination of the two pulses was measured with a Faraday imaging setup.

is detrimental for all-optical switching. Interestingly, the total threshold fluence for two-pulse switching was always higher than the one (σ)-pulse minimum threshold fluence. However, this might also be due to spot inhomogenities and a nonperfect spatial overlap.

In a second step we systematically decreased the fluence of the σ pulse. We observed that a lower threshold fluence exists, below which all-optical switching could not be realized anymore by adding the heating π pulse. In this case, a direct threshold from "no switching" to "pure thermal demagnetization" (helicity independent) was found. This indicates that some minimum circularity is needed for all-optical switching, thus excluding a pure thermal origin of the all-optical switching process.

In a further step we kept the fluence of both σ and π pulses constant and delayed them temporally. Figure 3 shows the

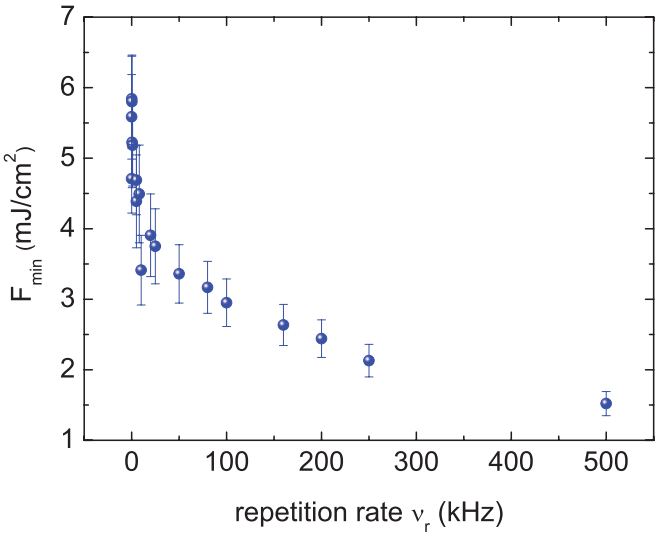

FIG. 2. (Color online) The minimum threshold fluence for all-optical switching decreases with increasing repetition rate. The repetition rate was varied from single shot to 500 kHz.

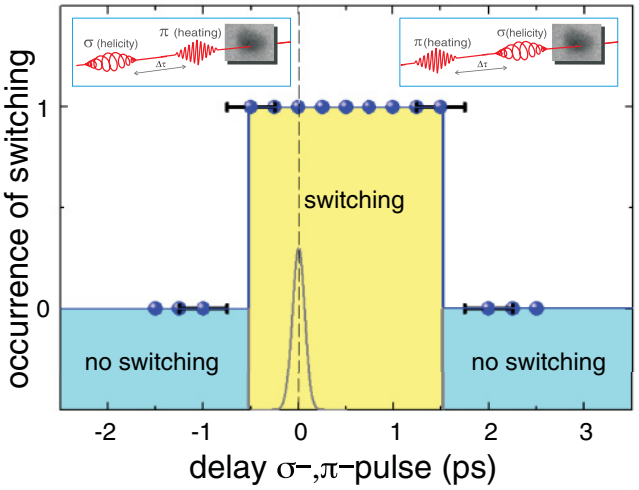

FIG. 3. (Color online) Results of the σ-π switching experiment, performed by varying the delay between the σ and the π pulse in both directions (measured at $\nu_r = 5.2$ kHz). Negative delays indicate that the π pulse arrives before the σ pulse, and positive delays vice versa. Occurrence of switching 1(0) means yes(no). We used $F_\pi = 1.93\text{mJ/cm}^2$ and $F_\sigma = 3.31\text{mJ/cm}^2$, which is lower than the threshold fluence for one-pulse switching ($F_{\text{min}} = 3.43\text{mJ/cm}^2$). The pulses had a FWHM of 100 fs, as sketched in the graph.

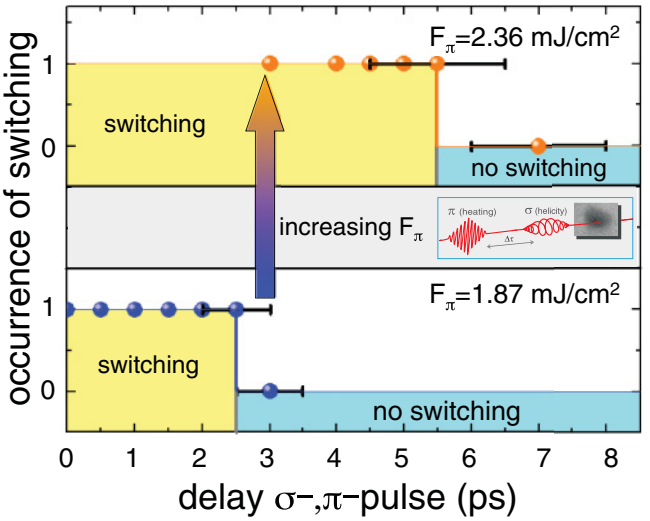

FIG. 4. (Color online) Systematical investigation of the maximum positive delay (meaning the σ pulse arrives first) for two-pulse switching (measured with pulse lengths of ≈700 fs and $\nu_r = 6.5$ kHz). F_σ was 4.96 mJ/cm^2 (lower than $F_{\text{min}} = 6.29$ mJ/cm^2) and kept constant. We started with $F_\pi = 1.87\text{mJ/cm}^2$ and increased it to $F_\pi = 2.36$ mJ/cm^2 after reaching the maximum positive delay where switching was still possible. Occurrence of switching 1(0) means yes(no).

delay range where two-pulse switching was obtained with one particular fluence set ($F_\pi = 1.93\text{mJ/cm}^2$, $F_\sigma = 3.31\text{mJ/cm}^2$, $FWHM = 100$fs). Negative delays indicate that the π pulse arrives on the sample before the σ pulse and vice versa for positive delays. As shown in Fig. 3 all-optical switching is possible for both delay signs. Additionally we found that the width of the delay window in which all-optical switching can be observed depends on the fluence of the π pulse. In particular, by increasing F_π this delay window can be extended. This is exemplarily demonstrated in Fig. 4 for two-pulse switching with fixed $F_\sigma = 4.96\text{mJ/cm}^2$ and variable F_π ($FWHM \approx 700$fs). For $F_\pi = 1.87\text{mJ/cm}^2$ (bottom) the switching window is about 3 ps wide, while for $F_\pi = 2.36$ mJ/cm^2 (top) we obtained around 6 ps. We performed the same experiment with different combinations of F_σ and F_π. We found that for $F_\sigma \geqslant F_\pi$ the delay window is some picoseconds wide, while if the π pulse is stronger than the σ pulse (on the order of the one pulse threshold fluence or more), then the resulting delay window is much wider, up to 100 ps. In this second case, it is still possible to write and delete all-optically, but multidomain switching is already observed at the central parts of the Gaussian spot profile.

The first conclusion that can be drawn from the results in Fig. 3 and Fig. 4 is the following: As we found an upper positive delay limit for two-pulse switching and the switching range can be extended only by increasing the heating fluence F_π, we can state that for a given amount of circularity (fixed F_σ) a certain amount of heat is really essential to switch all-optically. As a consequence, the classification of all-optical switching as a nonthermal effect that was proposed in Ref. 6 should be abandoned.

Let us now discuss the role of helicity for the switching process by comparing the results of the σ-π experiments for positive and negative delays. If the σ pulse arrives on the sample first (positive delay), the helicity information is imprinted on the system before it is sufficiently heated. Thus, we expect the positive delay window to be limited by the decay time of the helicity storage (τ_{hel}), at least for the case where τ_{hel} is shorter than the relevant heat diffusion time scale (τ_{heat}). If $\tau_{\text{heat}} < \tau_{\text{hel}}$, then the width of the positive delay window represents a lower limit for τ_{hel}. This brings us to the second conclusion: According to the data presented in Fig. 3 and Fig. 4 the decay time of the helicity storage in GdFeCo must be *at least* on the order of some picoseconds. We point out that for negative delays the σ pulse always brings "fresh" helicity information into the system. In this case, the width of the negative delay window cannot be limited by the decay time of the helicity storage.

To finally discern whether $\tau_{\text{heat}} < \tau_{\text{hel}}$ or vice versa, we performed an additional set of measurements where we chose a fluence pair (F_σ,F_π) to obtain a σ-π switching event, and then used the same fluence pair, but substituted the π pulse with a σ pulse (σ-σ switching, both pulses equally polarized). If $\tau_{\text{heat}} > \tau_{\text{hel}}$, then the width of the positive delay window should depend on whether the σ-σ or σ-π scheme is used. On the contrary, we observed no relevant changes, which implies that $\tau_{\text{heat}} < \tau_{\text{hel}}$. This means that the width of the positive delay window is effectively determined by τ_{heat} and thus constitutes a lower limit for τ_{hel}.

One possible candidate for helicity storage discussed in literature[30] are the Gd 4f moments. Hohlfeld *et al.* suggest that due to the strong hybridization of the d- and the low-lying f states, the f spins can store the helicity information although they cannot directly be optically excited and serve as nucleation points for the development of a switched domain. If this athermal model is correct, it should not be possible to switch at all if the fluence of the circularly polarized pulse is

below the one-pulse threshold. In contrast, our results favor a two-step-like process, where (1) the helicity is transferred to the material while the circularly polarized pulse is present ("input of the helicity"), and it is stored in a long-lived storage, and concomitantly (2) a certain amount of heating is needed to switch the sample magnetization.

But does the *input* of the helicity information into the material also depend on the temperature, in particular on the phonon temperature? A clue to answering this question follows from the fact that the switching window in Fig. 3 is asymmetric on the delay. Keeping in mind that $\tau_{\text{heat}} < \tau_{\text{hel}}$, the observed asymmetry can be understood by assuming that the *input* of the helicity information itself is temperature dependent. More precisely, the helicity input is enhanced for lower phonon temperatures. Accordingly if the σ pulse arrives when the phonon temperature is already strongly increased by the π pulse, insufficient helicity is transferred to the material to exceed the minimum circularity threshold.

We suggest that the temperature dependence of all-optical switching can be described by two competing effects: While the helicity *input* is enhanced if the sample is at low temperature, i.e., magnetically ordered, the switching process itself needs heating, i.e., magnetic disorder. In particular, the less helicity is transferred to the sample the more heating is necessary to switch. This idea might also explain the experimental data presented in Ref. 30. Here the laser fluence F_0 was set below the minimum threshold fluence determined at 300 K. Then the sample temperature was progressively decreased down to 10 K. For temperatures below 250 K all-optical switching could be achieved with the given fluence F_0. According to our findings, this behavior can be interpreted as follows: At 300 K the helicity transferred to the sample is not sufficient to exceed the minimum circularity needed to switch all-optically for given heating. In contrast, for temperatures below 250 K the helicity is brought into the sample more efficiently, and the circularity threshold can be overcome. Hence, for the lower temperatures switching is possible.

To conclude, we investigated the role of laser heating and helicity for all-optical switching in GdFeCo. We found that there is a very complicated interplay between these two factors. First, we could unambiguously show that heating contributes to the all-optical switching process, ruling out the classification of this process as a nonthermal effect. Second, we found that a minimum amount of circularity is needed to switch, excluding a pure thermal origin of all-optical switching. Third, we concluded that the helicity information of the exciting laser pulse is stored in the material for at least some picoseconds after optical excitation, and that the helicity input in the material is more efficient at lower temperatures.

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[34]The slight change of the pulse duration with repetition rate from 9 to 13 ps is not expected to have a significant influence on the switching threshold as shown in Ref. 27.