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Ultrafast Path for Optical Magnetization Reversal via a Strongly Nonequilibrium State

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Using time-resolved single-shot pump-probe microscopy we unveil the mechanism and the time scale of all-optical magnetization reversal by a single circularly polarized 100 fs laser pulse. We demonstrate that the reversal has a linear character, i.e., does not involve precession but occurs via a strongly nonequilibrium state. Calculations show that the reversal time which can be achieved via this mechanism is within 10 ps for a 30 nm domain. Using two single subpicosecond laser pulses we demonstrate that for a 5 μm domain the magnetic information can be recorded and readout within 30 ps, which is the fastest “write-read” event demonstrated for magnetic recording so far.

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The fundamental and practical limit of the speed of magnetization reversal is a subject of vital importance for magnetic recording and information processing technologies as well as one of the most intriguing questions of modern magnetism [1–8]. The conventional way to reverse the magnetization \mathbf{M} is to apply a magnetic field \mathbf{H} antiparallel to \mathbf{M} . In this collinear M-H geometry the reversal occurs via precession accompanied by damping that channels the associated angular momentum into the lattice. Although this process is perfectly deterministic, it is also unavoidably slow, typically of the order of nanoseconds, due to the required angular momentum transfer [8].

Alternatively, the driving field can be applied orthogonal to \mathbf{M} , so that the created torque $[\mathbf{M} \times \mathbf{H}]$ leads to a rapid change of the angular momentum and a possible switching of the magnetization [1,3,4,9]. However, such precessional switching requires a magnetic field pulse precisely tuned to half of the precession period. The fastest precessional reversal demonstrated so far using an external magnetic field [1,5] or a spin-polarized current [6,7,10] is limited to 100 ps. Moreover, it has been shown that for field pulses shorter than 2.3 ps such a switching becomes nondeterministic [5,11].

Ultrafast laser-induced heating of a magnetic material is known to stimulate the transfer of angular momentum from spins to lattice on a femtosecond time scale [12,13]. It has been recently demonstrated that a 40 fs, circularly polarized, laser pulse is able to reverse magnetization in a collinear M-H geometry [14], as if it acts as an equally short magnetic field pulse $\mathbf{H}_{\text{eff}} \sim [\mathbf{E} \times \mathbf{E}^*]$ (where \mathbf{E} is the electric field of light) pointing along the direction of light [15]. Although this experiment showed the intriguing possibility of triggering magnetization reversal with a subpi-

cosecond stimulus, the relevant time scales and mechanism of such an optically induced magnetization reversal are still unanswered questions, since a precessional switching within 40 fs would require enormous effective magnetic fields above 10^2 T and unrealistically strong damping. To address these questions we used femtosecond single-shot time-resolved optical imaging of magnetic structures and multiscale modeling beyond the macro-spin approximation. The combination of these advanced experimental and theoretical methods unveiled an ultrafast linear pathway for magnetization reversal that does not involve precession but occurs via a strongly nonequilibrium state.

In our experiments the amorphous ferrimagnetic 20 nm $\text{Gd}_x\text{Fe}_{100-x-y}\text{Co}_y$ films with perpendicular anisotropy [14] were excited by a single circularly polarized laser pulse (FWHM of about 100 fs, a central wavelength at $\lambda_0 = 800$ nm). A single linearly polarized probe pulse (FWHM = 100 fs, $\lambda_0 = 640$ nm) delayed with respect to the pump was used for ultrafast imaging of the magnetic domain structure by means of the magneto-optical Faraday effect. Magnetic domains with magnetization parallel (“up”) or antiparallel (“down”) to the sample normal are seen as white or black regions, respectively, in an image on a CCD camera [16]. After each “write-read” event, the initial magnetic state was restored by applying a magnetic field pulse. Taking images of the magnetic structure for different delays between the pump and probe pulses we were able to visualize the ultrafast dynamics of the laser-induced magnetic changes in the material.

Figure 1(a) shows images of magnetic domains in a $\text{Gd}_{24}\text{Fe}_{66.5}\text{Co}_{9.5}$ sample at different delays after excitation by right- (σ^+) or left-handed (σ^-) circularly polarized pulses. The images were obtained for both types of domains with initial magnetization up and down. In the first

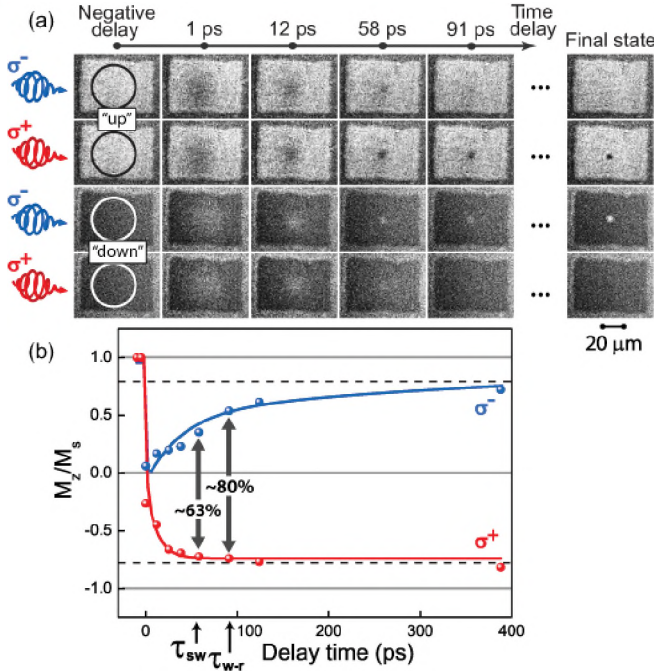


FIG. 1 (color). (a) The magnetization evolution in $\text{Gd}_{24}\text{Fe}_{66.5}\text{Co}_{9.5}$ after the excitation with σ^+ and σ^- circularly polarized pulses at room temperature. The domain is initially magnetized up (white domain) and down (black domain). The last column shows the final state of the domains after a few seconds. The circles show areas actually affected by pump pulses. (b) The averaged magnetization in the switched areas ($\sim 5 \mu\text{m}$) after σ^+ and σ^- laser pulses, as extracted from the images in (a) for the initial magnetization up.

few hundreds of femtoseconds, pump pulses of both helicities bring the originally magnetized medium into a strongly nonequilibrium state with no measurable net magnetization, seen as a gray area in the second column of Fig. 1(a), the size of which is given by the laser beam intensity profile. In the following few tens of picoseconds either the medium relaxes back to the initial state or a small ($\sim 5 \mu\text{m}$) domain with a reversed magnetization is formed. It is thus obvious that (i) the switching proceeds via a strongly nonequilibrium demagnetized state, clearly not following the conventional route of precessional motion, and (ii) the final state is defined by the helicity of the 100 fs pump pulse [last column of Fig. 1(a)].

As one can see from Fig. 1(a), the metastable state corresponding to reversed magnetization is reached within 60 ps after σ^+ (σ^-) excitation. This state is, however, slightly different from the final state [the last column in Fig. 1(a)], as clearly seen from Fig. 1(b). This happens due to the laser-induced heating of the sample followed by slow ($\gg 1$ ns) heat diffusion [17]. To take into account renormalization of the two metastable states of magnetization at the subnanosecond time scale we introduce two asymptotic levels [see dashed lines in Fig. 1(b)]. The characteristic time of switching τ_{sw} can be identified as the time required to reconstruct 63% ($1 - e^{-1}$) the difference between the

metastable states [Fig. 1(b)]. For example, in Fig. 1, $\tau_{sw} = 60$ ps. After $1.5\tau_{sw}$ the difference reaches 80% and, as also can be seen from Fig. 1(a), this time can be reliably assumed as the period required for a write-read event ($\tau_{w-r} = 90$ ps for the example in Fig. 1). The switching time is in fact surprising, because in contrast to heat-assisted magnetic recording [18], the reversal time is much longer than the effective light-induced magnetic field pulse H_{eff} . The duration of the latter Δt_{eff} is still an open question but can be different from the FWHM of the optical pulse. However, Δt_{eff} can be estimated from the spectrum of THz radiation generated by an Fe film when the latter is excited by a subpicosecond visible laser pulse. According to Ref. [19], the intensity of the THz emission depends on the polarization of the incoming light and has to be explained in terms of difference-frequency generation. Phenomenologically, this is very similar to the inverse Faraday effect. Based on a half-period oscillation with the lowest frequency in the THz spectrum [19], the maximum Δt_{eff} is about 3 ps. The pulse amplitude H_{eff} , for a typical pump fluence of 2.5 J/m^2 and the magneto-optical constant of GdFeCo ($\sim 3 \times 10^5 \text{ deg/cm}$), reaches 20 T.

To understand this route for magnetization reversal via such a strongly nonequilibrium state we solved the Landau-Lifshitz-Bloch (LLB) equation. This macrospin approach encapsulates very well the response of a set of coupled atomic spins subjected to rapidly varying temperature changes, including the reduction of the magnitude of \mathbf{M} [20,21]. The temperature dependence of the anisotropy constant K_u is introduced in the LLB equation via the temperature dependence of the transverse susceptibility [22]. The temperature-dependent parameters for the LLB equation, i.e., the longitudinal and transverse susceptibilities and the temperature variation of the magnetization, are calculated atomistically using Langevin dynamics combined with a Landau-Lifshitz-Gilbert equation for each spin [22]. It is well known that, due to the small heat capacity of electrons, optical excitation by a subpicosecond laser pulse can cause heating of the electron system well above 1000 K, whereafter the electrons equilibrate with the lattice to a much lower temperature on a (sub) picosecond time scale given by the electron-phonon interaction [13]. This laser-induced increase of the kinetic energy (temperature) of the electrons is simulated using a two-temperature model [23], the parameters for which were taken to be typical for a metal [24] (electron heat capacity $C_e = 1.8 \times 10^6 \text{ J/m}^3 \text{ K}$ at room temperature and electron-phonon coupling $G_{el-ph} = 1.7 \times 10^{18} \text{ J/K s}$). The simulations show that in the first 100 fs the electron temperature T_{el} increases from 300 K up to T_{el}^* and relaxes with a time constant of 0.5 ps down to the vicinity of T_C . Simultaneously the spins experience a short pulse of effective magnetic field with amplitude $H_{eff} = 20$ T and duration Δt_{eff} . The possibility of magnetization reversal under these circumstances has been analyzed numerically for a volume of $30 \times 30 \times 30 \text{ nm}^3$. The results of the simula-

tions are plotted in Fig. 2(a) as a phase diagram, defining the combinations of T_{el}^* and Δt_{eff} for which switching occurs for the given H_{eff} . The assumed perpendicular anisotropy value was $K_u = 6.05 \times 10^5 \text{ J/m}^3$ at 300 K. As can be seen from the diagram, a field pulse as short as $\Delta t_{\text{eff}} = 250 \text{ fs}$ can reverse the magnetization. For better insight into the reversal process we simulated the latter for $\Delta t_{\text{eff}} = 250 \text{ fs}$ and $T_{\text{el}}^* = 1130 \text{ K}$. The result is plotted in Fig. 2(b), showing that, already after 250 fs, the effective fields of two different polarities bring the medium into two different states, while the magnetization is nearly quenched within less than 0.5 ps. This is followed by relaxation either to the initial state or to the state with reversed magnetization, achieved already within 10 ps. The considered pulse duration Δt_{eff} of 250 fs is only 2.5 times larger than the FWHM of the optical pulse in our experiments [25] and well within the estimated lifetime of a medium excitation responsible for H_{eff} . Importantly, in simulations Δt_{eff} was found to be sensitive to the parameters of the two-temperature model. In particular, an increase of $G_{\text{el-ph}}$ leads to a reduction of the minimum field pulse duration. This shows that the suggested mechanism may, in principle, explain the experimentally observed laser-induced magnetization reversal. This magnetization reversal does not involve precession; instead, it occurs via a *linear* reversal mechanism, where the magnetization first vanishes and then reappears in a direction of H_{eff} , avoiding any transverse magnetization components, just as seen in Fig. 1(a). Exactly as in the experiments, the initial 250 fs effective magnetic field pulse drives the reversal process, that takes 1–2 orders of magnitude longer.

The state of magnetization after the pulse is critically dependent on the peak temperature T_{el}^* and the pulse duration. For ultrafast linear reversal by a 250 fs field pulse it

is necessary that, within this time, T_{el} reaches the vicinity of T_C . If, however, this temperature is too high and persists above T_C for too long, the reversed magnetization is destroyed and the effect of the helicity is lost. This leads to a phase diagram [Fig. 2(a)], showing that the magnetization reversal may occur in a certain range of T_{el}^* . Such a theoretically predicted reversal window of electron temperature can be easily verified in the experiment when one changes the intensity of the laser pulse. Figure 2(c) shows the switchability, i.e., the difference between the final states of magnetization achieved in the experiment with σ^+ - and σ^- -polarized pulses, as a function of T_{el}^* , calculated from the laser pulse intensity. It is seen that, indeed, switching occurs within a fairly narrow laser intensity range [26]. For intensities below this window no laser-induced magnetization reversal occurs, while if the intensity exceeds a certain level both helicities result in magnetization reversal, since the laser pulse destroys the magnetic order completely, which is then reconstructed by stray fields [27,28]. Such a good agreement between experiment and theory supports the validity of the proposed reversal mechanism.

Despite this qualitative agreement between simulations and experiments, the experimentally observed reversal time is several times larger than the calculated 10 ps. The latter, however, is calculated for a 30 nm domain, whereas in our experiments the magnetization in a $5 \mu\text{m}$ spot is manipulated. This size is defined by (i) the minimum size of the stable domain in the material and (ii) by the area within the laser spot, where the intensity favors the helicity-dependent reversal. Inhomogeneities in the sample and the intensity profile will lead to variations of T_{el}^* over the laser spot. If due to these factors every 30 nm element of the $5 \mu\text{m}$ spot is reversed with a probability between 50% and 100%, the actual time of magnetization reversal of this large spot will depend on its size and the speed of domain walls. Their mobility increases dramatically in GdFeCo alloys in the vicinity of their angular momentum compensation point (T_{comp}); i.e., the temperature where the angular momenta of the two sublattices cancel each other [29–31]. Therefore, one should expect a dramatic acceleration of magnetization reversal near T_{comp} . Note that this would also perfectly explain the difference between the times required for the formation of the switched domain and the relaxation to the initial state [Fig. 1(b)]. Indeed, in the former case the domain wall motion is additionally accelerated by the demagnetizing field, while in the latter case this field slows the motion down.

This hypothesis was verified experimentally by investigating the reversal process as a function of temperature in three alloys $\text{Gd}_{22}\text{Fe}_{68.2}\text{Co}_{9.8}$, $\text{Gd}_{24}\text{Fe}_{66.5}\text{Co}_{9.5}$, and $\text{Gd}_{26}\text{Fe}_{64.7}\text{Co}_{9.3}$ that are characterized by different compensation temperatures. The observed write-read time $\tau_{\text{w-r}}$ is plotted in Fig. 3 as a function of the difference between the sample temperature and the compensation point $T - T_{\text{comp}}$. The write-read time is the fastest and weakly de-

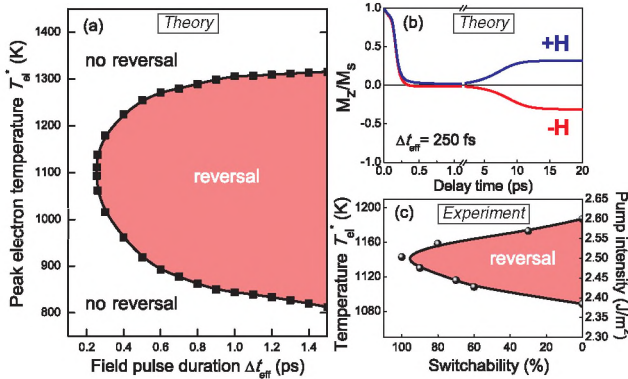


FIG. 2 (color). (a) Phase diagram showing the magnetic state of the $(30 \text{ nm})^3$ volume achieved within 10 ps after the action of the optomagnetic pulse with parameters $H_{\text{eff}} = 20 \text{ T}$, Δt_{eff} , and T_{el}^* . (b) The averaged z component of the magnetization versus delay time as calculated for 250 fs magnetic field pulses $H_{\text{eff}} = \pm 20 \text{ T}$ and $T_{\text{el}}^* = 1130 \text{ K}$. (c) Switchability versus the pump intensity for $\text{Gd}_{22}\text{Fe}_{68.2}\text{Co}_{9.8}$ at room temperature. We calculated the peak electron temperature T_{el}^* using C_e . Note that in this range of intensities the amplitude of the effective light-induced magnetic field varies within 19.2–20.8 T.

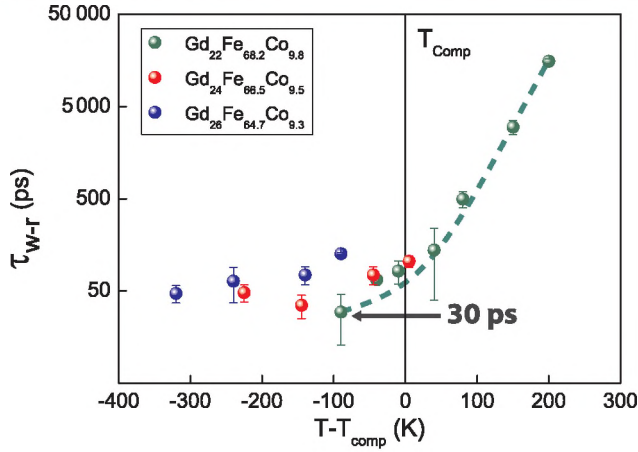


FIG. 3 (color). The write-read time τ_{w-r} versus the relative temperature $T - T_{\text{comp}}$ for $\text{Gd}_{22}\text{Fe}_{68.2}\text{Co}_{9.8}$ ($T_{\text{comp}} = 100$ K), $\text{Gd}_{24}\text{Fe}_{66.5}\text{Co}_{9.5}$ ($T_{\text{comp}} = 280$ K), and $\text{Gd}_{26}\text{Fe}_{64.7}\text{Co}_{9.3}$ ($T_{\text{comp}} = 390$ K). We achieved magnetization reversal within 30 ps for $\text{Gd}_{22}\text{Fe}_{68.2}\text{Co}_{9.8}$ at 10 K. The dashed line is guide to the eye.

depends on temperature below T_{comp} . This agrees with the hypothesis that the relaxation time to the metastable state is defined by the domain wall speed averaged over the photoexcited area. If the laser pulse brings the central part of the excited area from initial temperature $T < T_{\text{comp}}$ to the vicinity of T_C , somewhere within this area the material is at T_{comp} , where the domain wall mobility is the largest. Then, it is the mobility at T_{comp} which dominates the averaged domain wall speed in the photoexcited area and, thus, determines the write-read time. Above T_{comp} , all-optical magnetization reversal can still be realized, but the write-read time increases exponentially with increasing temperature. For example, while τ_{w-r} for $\text{Gd}_{22}\text{Fe}_{68.2}\text{Co}_{9.8}$ ($T_{\text{comp}} = 100$ K) at room temperature is found to be extremely slow (16 ns), a huge decrease of τ_{w-r} of 2 orders of magnitude is observed as $T - T_{\text{comp}}$ decreases. Finally, at 10 K we succeeded to achieve all-optical magnetization reversal within just 30 ps, which is the fastest write-read event demonstrated for magnetic recording so far.

In conclusion, by time-resolved single-shot microscopy, we found a novel and ultrafast path for magnetization reversal triggered by a subpicosecond circularly polarized laser pulse. The reversal does not involve precession, but instead has a linear character, proceeding via a strongly nonequilibrium state. This all-optical reversal occurs only in a narrow range of pulse energies. Using two single subpicosecond laser pulses we demonstrated the feasibility of both all-optical recording and reading on an ultrashort time scale. The magnetic information was recorded by a subpicosecond laser pulse in a $5 \mu\text{m}$ domain and readout by a similarly short pulse within 30 ps, which is the fastest write-read event demonstrated for magnetic recording so far. Simulations for 30 nm domains demonstrate the feasibility of reversing magnetization within 10 ps. This time can be even faster for media with a higher magnetic anisotropy constant than the one used in our calculations.

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