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ABSTRACT

All-optical switching of magnetic order presents a promising route toward faster and more energy efficient data storage. However, a realization in future devices is ultimately dependent on the maximum repetition rates of optically induced write/erase cycles. Here, we present two strategies to minimize the temporal separation of two consecutive femtosecond laser pulses to toggle the out-of-plane direction of the magnetization of ferrimagnetic rare-earth transition metal alloys. First, by systematically changing the heat transfer rates using either amorphous glass, crystalline silicon, or polycrystalline diamond substrates, we show that efficient cooling rates of the magnetic system present a prerequisite to accelerate the sequence of double pulse toggle switching. Second, we demonstrate that replacing the transition metal iron by cobalt leads to a significantly faster recovery of the magnetization after optical excitation allowing us to approach terahertz frequency of write/erase cycles with a minimum pulse-to-pulse separation of 7 ps.

Research of ultrafast light control of magnetic order[1,2] is driven by the quest to find faster and more energy efficient platforms to store and manipulate information. Certainly, the most remarkable observation in this research field is the deterministic and ultrafast magnetization reversal upon femtosecond optical excitation in metallic GdFe alloys.[3,4] In spite of extensive research, only a very limited number of magnetic systems have been shown to exhibit single pulse all-optical switching (AOS): These include different compositions of metallic ferrimagnetic GdFeCo,[3,5] GdCo[6] and GdTbCo[7] amorphous alloys, Tb/Co multilayers,[8] synthetic Gd-transition metal heterostructures,[9,10] the half-metallic ferrimagnetic Heusler alloys Mn_2Ru_xGa,[11] and the optically transparent Cobalt-substituted yttrium iron garnet.[12] A very different approach to optically induce changes in magnetization is based on reversible structural changes in phase change materials, such as FeRh[13,14] or Fe(Co)-based alloys,[15,16] and is attracting considerable attention for data storage applications. Only very recently, however, research has started to address the important question, which mechanisms limit the repetition rate of optically driven write/erase cycles. While a theoretical study predicts terahertz repetition rates,[17] much larger than required for equilibration of the involved subsystems of electrons, spin and phonons, an extensive experimental work investigating the double-pulse magnetization dynamics in a $Gd_{27}Fe_{64}Co_9$ alloy revealed a minimum separation of the two pulses of $\Delta t_{12} = 300$ ps for reliable, consecutive toggle switching.[18] However, guided by heat diffusion calculations, they predicted a potential increase in achievable repetitions rates, either via improved heat sinks and, therefore, accelerated cooling rates or by tailoring the composition of the alloy in order to reduce the required fluence threshold for AOS, i.e., the minimum laser fluence to induce deterministic switching. Interestingly, a double pulse experiment on the Heusler alloy Mn_2Ru_xGa grown on a crystalline MgO substrate revealed much shorter minimal time intervals between two consecutive pulses, dropping as a function of excitation fluence down to $\Delta t_{12} = 12$ ps.[19]

Here, we follow two strategies to increase the repetition rate of all-optical magnetic write/erase cycles: First, we compare the dual-pulse excitation dynamics for a GdFe alloy on three different substrates characterized by vastly different heat conductivities. For amorphous glass, we measure a Δt_{12} in the ns regime and for

crystalline silicon as well as for polycrystalline diamond $\Delta t_{12} \leq 500$ ps. Second, motivated by the very fast switching dynamics recently observed in GdCo alloys,[6,7] we investigated the response of a Si/GdCo system and demonstrate reliable write/erase cycles with $\Delta t_{12} = 7$ ps, corresponding to a frequency of 140 GHz.

All measurements were performed using a wide-field magneto-optical microscope with femtosecond temporal and sub-micrometer spatial resolution as recently described by us.[20] A Yb-based fiber laser with a pulse length of 250 fs and a central wavelength of 1030 nm is used as the light source. Each of the optical paths of the two excitation pulses is equipped with a separate lambda-half plate and a polarizer to accurately control their respective pulse energies. Both pump pulses are aligned collinearly and focused by the same lens onto the sample with an incident normal angle of 14°. The size of the resulting, slightly elliptical footprint, is determined via a calibrated beam profiling camera and allows us to determine accurate values of the two excitation fluences, F_1 and F_2. Note that all given fluence values refer to incident fluences. The time resolved experiments are performed in a Faraday geometry: We split off a fraction of the laser pulse, half its wavelength in a non-linear crystal to 515 nm, and illuminate the sample in a transmission geometry. In the time-resolved single pulse experiments, we use an external, out-of-plane magnetic field to reset the magnetization after every laser shot. Final-state, magneto-optical images after single or double pulse excitation are recorded in Kerr geometry using an incoherent light emitting diode with the same probe wavelength of 515 nm. All double-pulse all-optical switching experiments are performed without any external magnetic field. The reflected or transmitted light of the samples is projected onto an active-pixel sensor by a long distance objective and a tube lens. The magneto-optical contrast emerges in a crossed-polarizer geometry, where an analyzer optic behind the sample is set close to 90° with respect to the direction of the incoming light polarization. The temporal separation Δt_{12} of the two excitation pulses is controlled via a mechanical delay stage; for values exceeding 1 ns, we delay the second pulse in steps of 1 ns by folding the beam path via mirror-pairs. We fix the fluence of the first pulse, F_1 at 0.5 mJ/cm^2 above the fluence threshold of AOS and scan the fluence of the second pulse, F_2, for every value of Δt_{12}. Importantly, as the magnetization depends nonlinearly on the detected intensity, we determine the ultrafast evolution between the initial and final magnetization state by extracting the Faraday angle at each time step via automated analyzer scans.[20] A sketch of the setup is depicted in Fig. 1(a).

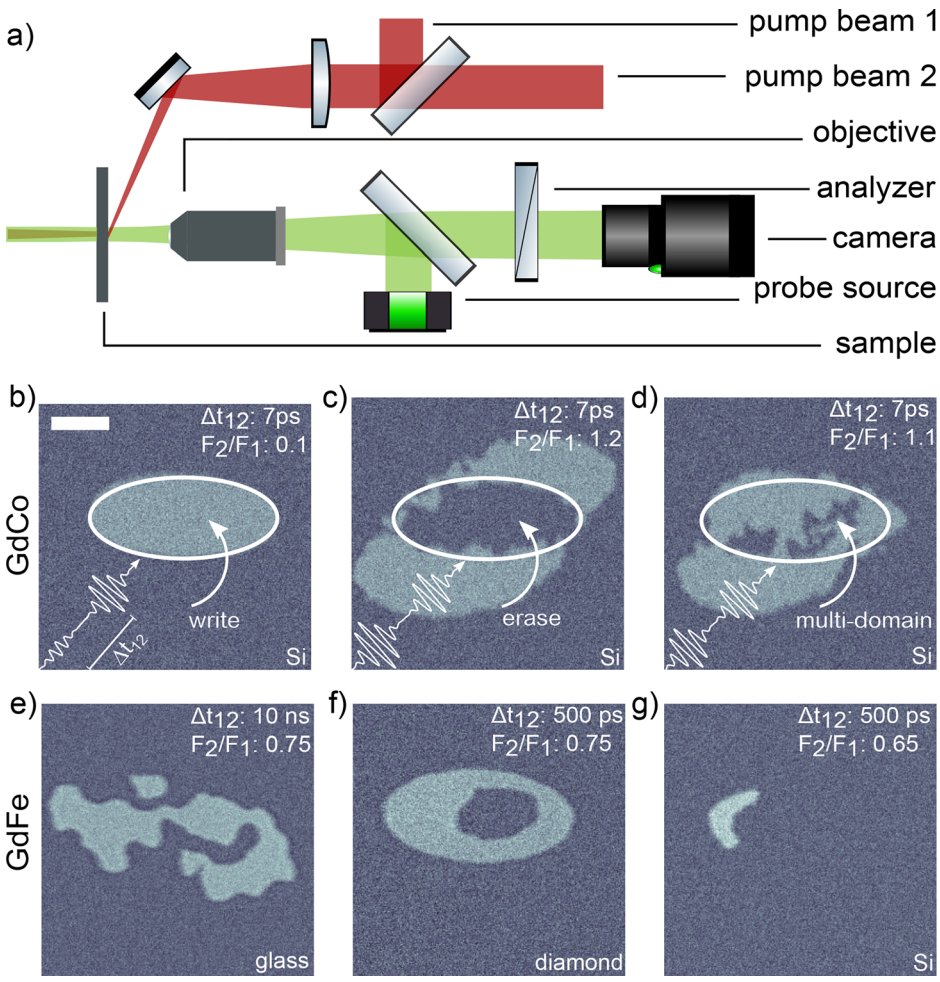

FIG. 1. (a) Faraday/Kerr Microscope to investigate double-pulse AOS. Final state, magneto-optical images of Si/GdCo at $\Delta t_{12} = 7$ ps for (b) $F_2/F_1 = 0.1$ displaying single AOS, (c) $F_2/F_1 = 1.2$ displaying double AOS for as well as for (d) $F_2/F_1 = 1.1$ displaying a multi-domain state. GdFe sample on (e) glass exhibiting a multi-domain state for all values of $\Delta t_{12} < 10$ ns and on (f) diamond and (g) and Si exhibiting double AOS for $\Delta t_{12} > 500$ ps. The scale bar is 10 μm.

The geometry and composition of the investigated samples are as follows: first, substrate/Ta(3 nm)/$Gd_{24}Fe_{76}$(20 nm)/Ta(3 nm), for the following substrates: amorphous glass, crystalline silicon, and polycrystalline diamond, second substrate/Ta(3 nm)/Pt(3 nm)/$Gd_{22}Co_{78}$(20 nm)/Ta(3 nm), where the substrate is either amorphous glass or crystalline silicon. Both samples are grown by magnetron sputtering and exhibit an out-of-plane magnetic anisotropy with square hysteresis loops, leading to two stable magnetization states with opposite out-of-plane directions. Both samples are below their compensation point at room temperature.

In Fig. 2, we compare the ultrafast evolution of single pulse AOS for a glass/GdFe and a glass/GdCo alloy for the same excitation fluence of 4.5 mJ/cm^2. Note that the time axis is linear up to 10 ps and logarithmic for larger time delays up to 1000 ps. A complete fluence-delay map of the GdCo sample can be found in the supplementary material. Within our time resolution of ≈300 fs, we find identical ultrafast demagnetization dynamics for GdFe and GdCo. However, the switching dynamics is apparently very different: While for GdCo the magnetization reaches a reversed magnetization of $M(7\,\mathrm{ps})/M(t<0) = -0.6$, it takes almost 30 times longer to reach the same value in GdFe. We expect that the relaxation time after single pulse excitation ultimately limits the frequency of write/erase cycles.

In Figs. 1(b)–1(g), we display final-state, magneto-optical images after double-pulse excitation for the Si/GdCo as well as for the GdFe sample grown on glass, silicon, and diamond. We define three different magnetization states: (i) a single AOS state, when the second pulse is too weak to influence the switching induced by the first pulse (panel c), (ii) a double AOS state, when the second pulse resets the magnetization back to its initial value, i.e., erases the bit written by the first pulse [panels (b), (f), and (g)]. Note that the area of the reset magnetization state is controlled by the fluence ratio as well as the exact spatial overlap of both pulses. Finally, (iii) a magnetic multidomain state, when the second pulse destroys the initially switched state leading to a random distribution of magnetic domains with opposite direction within the excited region [panel (d) and (e)].

The corresponding phase diagrams of Si/GdFe and diamond/GdFe as a function of Δt_{12} and F_2/F_1 are displayed in Fig. 3. We fix $F_1 = 7.3$ and $F_1 = 5.9$ mJ/cm^2 for the Si/GdFe and diamond/GdFe samples, respectively. The region for which we observe a double AOS state increases for a larger temporal separation of the two excitation pulses and shifts to higher values of the fluence ratio. For the diamond substrate, the window is significantly larger and re-switching requires a larger fluence of the second pulse. For both samples, the minimum value of the pulse-to-pulse separation is between $300\,\mathrm{ps} < \Delta t_{12} < 500$ ps. For the glass/GdFe sample, we find no reliable re-switching for values of up to $\Delta t_{12} = 10$ ns. A complete overview of the real space magneto-optical images of the final state after double pulse excitation can be found in the supplementary material. To reach a better understanding of the cooling effect of the different substrates, we performed one dimensional heat diffusion calculations based on a two-temperature model describing the response of electrons and lattice after optical excitation.[21] Upon absorption of the femtosecond laser pulse, the electronic temperature increases to above 2000 K, a value significantly exceeding the Curie temperature of the ferrimagnetic GdFe or GdCo alloys. Via strong electron-phonon scattering, the electronic and phononic systems equilibrate within ≈1.5 ps while further cooling of the lattice on longer time scales is strongly influenced by the thermal properties of the substrates. In Fig. 4, we display the calculated electron and lattice temperature of the GdFe or GdCo alloy as a function of time. We emphasize that the different incident threshold fluences for glass (4.9 mJ/cm^2), silicon (7.3 mJ/cm^2), and diamond

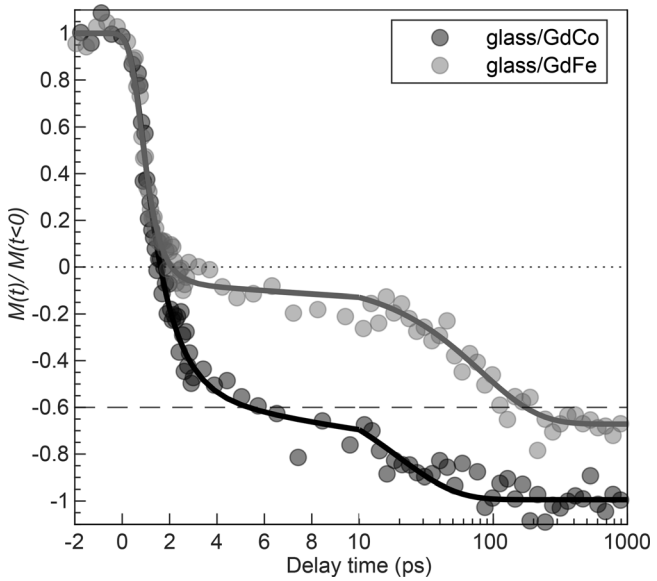

FIG. 2. Relative change of the magnetization, $M(t)/M(t<0)$, of glass/GdCo and glass/GdFe as a function of delay time. Note that the time axis is linear up to 10 ps and logarithmic between 10 and 1000 ps. GdCo exhibits a very fast relaxation toward a reversed magnetization reaching $M(7\,\mathrm{ps})/M(t<0) = -0.6$, whereas GdFe reaches this value only after ≈200 ps.

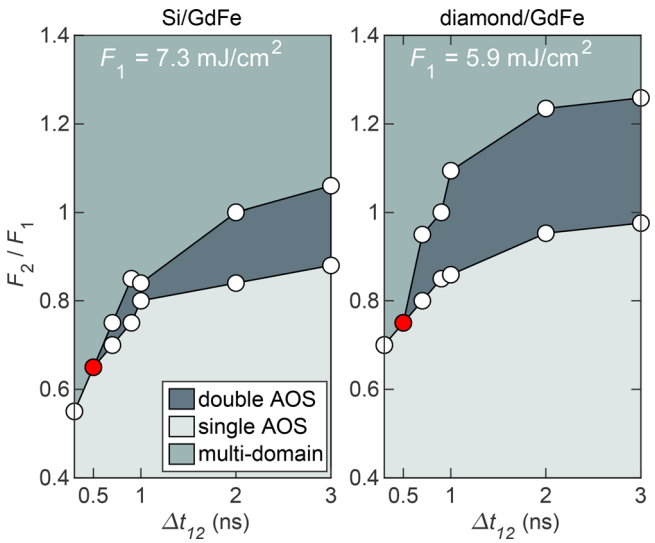

FIG. 3. Phase diagram of Si/GdFe and diamond/GdFe for double pulse excitation as a function of the fluence ratio F_2/F_1 and of the pulse-to-pulse separation Δt_{12}. For the diamond substrate, we observe a broader region of re-switched states. For both samples, the critical value for reliable write/erase cycles is $\Delta t_{12} = 500$ ps (marked with a red dot).

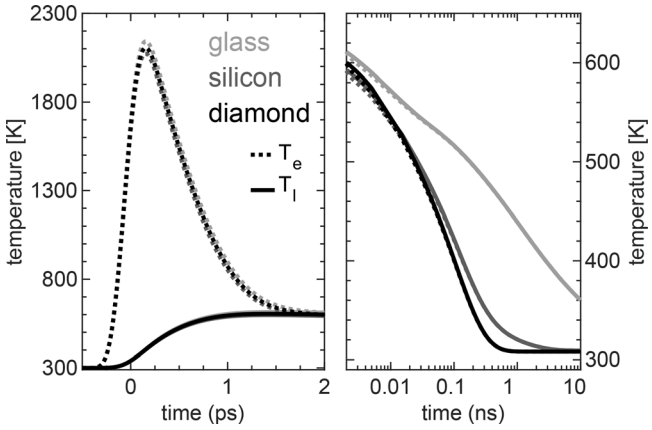

FIG. 4. Sample averaged electron, T_e, and lattice, T_l, temperature of the ferrimagnetic GdFe or GdCo alloy on glass, silicon, and diamond substrate as a function of time after single pulse excitation. The left panel shows the early dynamics on a linear time axis, the slower dynamics up to 10 ns is shown on a logarithmic scale.

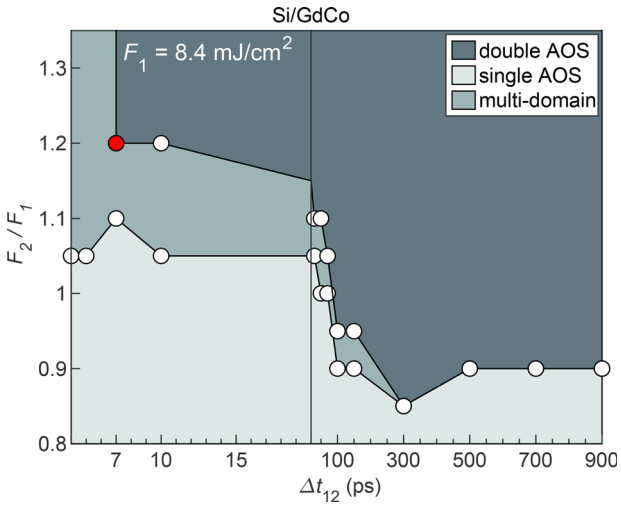

FIG. 5. Phase diagram of Si/GdCo for double pulse excitation as a function of the fluence ratio F_2/F_1 and of the pulse-to-pulse separation Δt_{12}. The critical value for reliable write/erase cycles is $\Delta t_{12} = 7$ ps (marked with a red dot). Note that for early times, reliable re-switching relies on an increased fluence of F_2.

(5.9 mJ/cm^2) are related to the different refractive indices of the substrates resulting in a nanostructure-specific absorption of GdFe but lead to almost identical electronic temperatures of GdFe. We note that the importance of the specific geometry of nanoscale layer systems for determining the effective absorption of the magnetic film is often overlooked in literature and may be one reason for the variation of reported threshold fluences.[5] As expected, the poor heat conductivity of amorphous glass results in a very slow reduction of the lattice temperature, while for diamond and silicon temperatures of ≈300 K are reached again after ≈500 ps. Our finding that the choice of substrate is crucial for high repetition rate toggle switching is plausible, as it has been shown already some time ago that reliable AOS takes place most efficiently for temperatures in the vicinity of the compensation temperature.[22–25] Note that this is also in agreement with the interpretation of Wang *et al.*,[18] who postulate a required cooling of the magnetic sample close to its compensation temperature before re-switching can take place. Evidently, an efficient heat transfer out of the magnetic sample is a prerequisite to reach competitive switching speeds. However, the comparable minimum pulse-to-pulse separation of <500 ps observed for the silicon and diamond substrates, in spite of their very different heat conductivities, suggests that the intrinsic single pulse switching dynamics of GdFe sets a lower bound for Δt_{12}.

To investigate this further, we turn to the GdCo alloy. In Fig. 5, we display the corresponding phase diagram for the Si/GdCo system for a fixed value of $F_1 = 8.4$ mJ/cm^2. Interestingly, we observe a very different response compared to the GdFe alloys. While for low values of F_2, the system remains in the state set by the first pulse (single AOS), increasing F_2 first leads to a narrow region of multi-domain magnetic states, before reliable double AOS occurs. Most notably, the minimum value of Δt_{12} is reduced by two orders of magnitude compared to Si/GdFe and amounts here to 5 ps $< \Delta t_{12} <$ 7 ps for a fluence ratio $F_2/F_1 \geq 1.2$. We further note that this very small value for the minimal required pulse-to-pulse separation is apparently not related to a reduced single pulse fluence threshold for AOS, a potential strategy that has been suggested to reach higher write/erase repetition rates.[18] For the glass/GdCo system, we do not observe any re-switching for $\Delta t_{12} \leq 900$ ps (see the supplementary material for the corresponding phase diagram of glass/GdCo as well as for a table of magneto-optical images for Si/GdCo as a function of F_1/F_2 and Δt_{12}).

There are two interesting aspects that are worth a further discussion: First, why does GdCo relax so much more efficiently to its reversed magnetization compared to GdFe and second why does the re-switching process require a higher energy of the second pulse? AOS has been rationalized by a phenomenological model[26,27] characterized by three distinct regimes: First, while the electron temperature is increased after impulsive laser excitation, the two antiferromagnetically aligned sublattices are decoupled and relativistic relaxation leads to distinct demagnetization rates proportional to the respective atomic magnetic moments. Furthermore, it was recently realized that such distinct demagnetization rates can also be influenced by element-specific damping, i.e., depend on the element-specific spin–orbit coupling strength.[28] Then, provided a suitable excitation fluence is set, the transition metal Fe or Co is fully demagnetized, while the rare earth element Gd still exhibits a finite magnetic moment. This marks the transition to the second regime dominated by exchange relaxation.[29] Now the total angular momentum is assumed to be a conserved quantity and the dynamics is characterized by transfer of angular momentum between the sublattices. The spin moment of Co increases at the expense of Gd and, because of their antiparallel spin direction, Co spins reverse and a transient ferromagnetic-like state emerges.[4] In the final step, relaxation to a reversed magnetic moment with antiferromagnetic alignment proceeds again via coupling to the external bath. Microscopically, one may rationalize the observed accelerated relaxation of the Co sublattice during the last step of AOS due to a stronger Co–Co exchange interaction compared to Fe–Fe exchange interaction in ferrimagnetic GdFe and GdCo alloys.[30] We argue that the exchange interaction of the transition metal will dominate the early relaxation dynamics. First of all, it by far exceeds the Gd–Gd as well as the Gd–Co/Fe interaction energy[31,32] and additionally most nearest neighbor pairs are formed by transition metal atoms for the stoichiometry

of our sample. We emphasize that this notion is also supported by a recent theoretical work[33] that established criteria for the transitions between different relaxation regimes during AOS. Here, increasing the inter-atomic exchange interaction within the transition metal sublattice is predicted to accelerate the changeover from exchange to relativistic relaxation leading to a speed up of the switching dynamics for GdCo vs GdFe. For longer times scales exceeding ≈150 ps, the different magnetization dynamics of GdFe and GdCo dynamics can be rationalized by their different Curie temperatures. We estimate values of ≈570 K for GdCo and ≈515 K for GdFe by a combined analysis of our time resolved measurements shown in Fig. 2, the heat diffusion calculation shown in Fig. 4 and phenomenological relations for $M(T)$. These values of T_C are in good agreement with literature values[7,31,34] (for more details we refer to the supplementary material).

The observation that for short separations of the two excitation pulses, Δt_{12}, GdCo requires a higher fluence of the second pulse to promote re-switching is currently not well understood. In line with the above arguments regarding the dominating role of the Co–Co exchange interaction for early, picosecond relaxation times, we assume a transient non-equilibrium, inter-sublattice distribution of magnetic moments, i.e., Co atoms magnetically order more quickly than Gd atoms.[33] Starting from this non-equilibrium state, the fluence of the second laser pulse, F_2, determines whether the sample enters the exchange relaxation regime with appropriate sublattice moments for AOS to take place. We speculate that for $F_2/F_1 < 1$ the Co moment is not fully quenched such that exchange relaxation never dominates the dynamics. For $F_2/F_1 \approx 1$, similar sublattice magnetic moments are induced, resulting in no preferential direction of angular momentum flow and the emergence of a multi-domain pattern. Only for $F_2/F_1 > 1$, the Gd moment is still finite as the moment of Co approaches zero such that intersublattice exchange relaxation leads to re-switching. Clarifying these questions and hypotheses clearly calls for further experimental studies, ideally providing element-specific information of both sublattices.

In conclusion, we have experimentally demonstrated the importance of an efficient cooling rate via optimized substrates in order to accelerate double pulse all-optical switching dynamics in ferrimagnetic GdFe and GdCo alloys. We find that the intrinsic switching dynamics of the ferrimagnetic magnetic system itself sets a lower limit for the highest achievable repetition rates. For a GdCo alloy exhibiting a much faster relaxation to a reversed magnetic state, we show reliable re-switching for a pulse-to-pulse separation of 7 ps, approaching terahertz repetition rates for write/erase cycles of magnetic bits.

See the supplementary material for an overview of the final state magneto-optical images both as a function of F_2/F_1 and Δt_{12} for the Si/GdFe and Si/GdCo sample. Furthermore, for the glass/GdCo sample, we show the magnetization dynamics as a function of time and fluence as well as its phase diagram for double pulse excitation. Finally, we provide additional details on how to extract approximate values of T_C for the GdFe and GdCo sample.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

DATA AVAILABILITY

The data that support the findings of this study are openly available in Zenodo at 10.5281/zenodo.6349558, Ref. 35.

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