Ultrafast Helicity-Independent All-Optical Switching in Amorphous (Gd,Tb)Co Thin Films

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Ultrafast control of the magnetization in ps timescales by fs laser pulses offers an attractive avenue for applications such as fast magnetic devices for logic and memory. However, ultrafast helicity-independent all-optical switching (HI-AOS) of the magnetization has thus far only been observed in Gd-based, ferrimagnetic rare earth-transition metal (RE-TM) systems, and a comprehensive understanding of the reversal mechanism remains elusive. Here, we report HI-AOS in amorphous (a-), ferrimagnetic a-Gd22−xTb30 thin films, from x = 0 to x = 18, and elucidate the role of Gd in HI-AOS in RE-TM alloys and multilayers. Increasing Tb content results in slower remagnetization rates and higher critical fluences for switching. Simulations of the atomistic spin dynamics based on the two-temperature model reproduce these results qualitatively and predict that the low damping on the RE sublattice arising from the small spin-orbit coupling of Gd (with L = 0) is instrumental for the faster dynamics and lower critical fluences of the Gd-rich alloys. Increasing the RE damping on a-Gd10Tb20Co78 sublattice by annealing leads to slower dynamics, confirming this prediction. These simulations strongly indicate that taking account of element specific damping, rather than a net damping of the system, is crucial in understanding HI-AOS phenomena, and the results suggest that engineering the element specific damping of materials can open up new classes of materials that exhibit low-energy, ultrafast HI-AOS.

The ability to control magnetism at short ps and sub-ps timescales has tantalized scientists since the discovery in 1997 of the ultrafast demagnetization of Ni following irradiation by fs laser pulses, opening up the field of ultrafast magnetization dynamics. A major breakthrough towards this end has been the discovery of helicity-independent all-optical switching (HI-AOS) of the magnetization, sometimes referred to as thermally-induced magnetization switching (TIMS), in ferromagnetic GdFeCo alloys in ps timescales by a single fs laser pulse. The high switching speed associated with HI-AOS offers the possibility of promising technological applications in high-speed, energy-efficient and non-volatile magnetic memory and logic, with two or three orders of magnitude of higher operating speed compared to conventional spintronic devices that operate on mechanisms such as external field control, spin-transfer torque or spin-orbit torque.

Despite significant advances in the field, a complete understanding of the mechanism of HI-AOS still remains lacking. Thus far, deterministic ultrafast toggle switching of the magnetization by a single laser pulse – where the switched area is controlled just by heating from the spatial distribution of the laser pulse fluence – has only been reported in Gd-based RE-TM ferrimagnetic alloys. These include GdFeCo, GdCo/Pt/Co/Gd and exchange coupled Co/Pt/Co/GdFeCo systems. Similar Tb-based ferrimagnetic RE-TM alloys and multilayers such as TbCo and Tb/Co multilayers have so far only shown helicity-dependent AOS (HD-AOS), requiring multiple circularly polarized laser pulses over longer timescales (µs to ms) or transient reversal with a single laser pulse, wherein the magnetization reverts back to its original direction after a short reversal of a few ps. Single-shot AOS was demonstrated in TbFeCo alloys but it required patterning of nanoscale antennas to enhance the optical field, thereby confining the switched region to less than 100 nm in areas near and around the antennas. The switching was strongly influenced by inhomogeneities and control of the uniformity in the switched area under the laser pulse fluence profile could not be achieved. Despite the progress in the field, a complete microscopic theory has yet to explain the intrinsic fundamental physics of the phenomenon, including addressing the apparent exclusivity of Gd in enabling HI-AOS in Gd-TM alloys and multilayers.

In this work the role of Gd in enabling HI-AOS was in-
FIG. 1. a) MOKE microscopy images illustrating samples with ability to all-optically reverse their magnetization upon irradiation. Samples with a Tb concentration of up to 18% exhibited HI-AOS while α-Tb22Co78 only exhibited demagnetization as evidenced by the nucleation of random domains. b) Intrinsic anisotropy constant $K_{ui}$ vs Tb atomic percent in α-Gd18Tb4Co78 thin films. Anisotropy increases with increasing Tb content but no correlation was found between HI-AOS and anisotropy. The green box shows the compositions that exhibited HI-AOS; the red box those that did not. The effect of growing on different substrates and capping layers was tested but it had no effect on HI-AOS. Three separate α-Gd-Co samples are shown. At the top is α-Gd19Co81 with compensation T below RT exhibiting PMA and HI-AOS. Beneath it is α-Gd22Co78 which is part of the main study. In red is α-Gd22−xTbxCo78 which has in-plane magnetization which cannot be probed using polar MOKE, hence it’s unknown if it exhibits HI-AOS. c) Incident and absorbed critical fluence increase with increasing Tb content.

investigated experimentally and theoretically by studying α-Gd22−xTbxCo78 thin films. By systematically replacing the Gd atoms with Tb atoms such that the RE composition is kept constant, as well as by post-growth annealing, the magnetization, anisotropy, magnetic damping and spin-orbit coupling are modified, permitting the study of their effects on the ultrafast magnetization dynamics and their influence on HI-AOS. HI-AOS was found in α-Gd22−xTbxCo78 films of up to x = 18, while α-Tb22Co78 only demagnetized upon laser irradiation. The threshold or critical fluence of the pulse needed for switching increased linearly with increasing Tb concentration and the switching dynamics became slower, indicating increasing difficulties in the switching process with Tb. Simulations of the atomistic spin dynamics using the VAMPIRE software package combined with a two-temperature model (2TM) indicate that these results can be explained by an increased damping of the RE site when Gd is replaced by Tb owing to the larger spin-orbit coupling in Tb due to its $L = 3$ value compared to $L = 0$ in Gd. The lower element specific damping of the Gd sublattice contributes to the lower critical fluences and faster switching speeds of the Gd-rich alloys. Slower dynamics in α-Gd10Tb12Co78 were observed after annealing at 300 °C for one hour, which is expected to increase the damping of the system, and the slower dynamics are reproduced by increasing the Gd damping in the simulation. From the simulations we observe that taking into account the individual elemental damping of the sublattices is crucial in reproducing the HI-AOS behavior observed. The results and simulations suggest that engineering the relative element specific damping of the RE and TM sublattices can help unravel new materials that exhibit HI-AOS and will help elucidate the mechanism behind HI-AOS and other ultrafast magnetism phenomena. Increased anisotropy with Tb content, and decreased anisotropy by annealing both led to slower switching dynamics, indicating that anisotropy is not a significant factor in HI-AOS. However, the high perpendicular magnetic
anisotropy (PMA) in Tb-rich films make them attractive candidates for magnetic devices with higher memory storage densities and retention times wishing to exploit the fast read-write speeds of HI-AOS.

RESULTS

Sample growth

Amorphous, ferrimagnetic thin-films of Ta(3)/Pt(3)/a-Gd$_{2x-}$Tb$_{78}$Co(10)/Pt(3) (thicknesses are in nm) heterostructures were sputter deposited onto substrates of Si(525 µm)/SiO$_2$(50 nm)/Si$_3$N$_4$(300 nm) by co-depositing from separate Tb, Gd and Co targets (see methods for sample growth and characterization details). Energy dispersive spectroscopy images taken with a scanning transmission electron microscope found no evidence of inhomogeneities at the 10 nm scale as reported in GdFeCo$_{22}$-Gd$_{78}$-(50nm)/SiN$_x$(300 nm) by co-depositing from separate Tb, Gd and Co targets (see methods for sample growth and characterization details). The magnetization was measured with a SQUID magnetometer and found the compensation temperature ($T_M$) of all samples to be near 400 K due to the fixed 22 at.% RE content, allowing to maintain nearly constant values of saturation magnetization ($\sim$ 100 emu/cc) and $T_M$ while varying the Gd/Tb ratio. At room temperature, the magnetization of all the films studied is RE-dominant. The Curie temperature of all films was found to be greater than 600 K (see methods).

Single-shot HI-AOS in a-GdTbCo alloys

The magnetization of the samples is initialized with an external out of plane magnetic field of $\sim$ 0.7 T, fully saturating all samples. Samples are then irradiated with 100 fs full-width half maximum (FWHM) optical pulses from a regeneratively amplified Ti-Sapphire laser (see methods). Magneto Optical Kerr Effect (MOKE) microscope images depict single shot switching of the magnetization across all films except Tb$_{22}$Co$_{78}$ as shown in Fig. 1a. It is seen that films with as little as 4 at.% Gd have deterministic magnetization reversal upon irradiation with a single laser pulse. Amorphous Tb$_{22}$Co$_{78}$ only shows demagnetization as evidenced by the nucleation of random magnetic domains.

The intrinsic anisotropy constant ($K_{in}$) was measured at room temperature and is plotted as a function of Tb at.% in Fig. 1b. The anisotropy constant increases systematically with increased Tb due to its large single ion anisotropy contribution. The effect of growing on Ta or SiN, and of capping with Ta or Pt are also shown in Fig. 1b; neither buffer nor capping layer tested had any impact on a film’s ability to switch via HI-AOS.

Fig. 1c shows that increasing the Tb content increases the incident critical fluence, starting from 4.4 mJ/cm$^2$ for a-Gd$_{22}$Co$_{78}$ and linearly increasing to 6.2 mJ/cm$^2$ for a-Gd$_4$Tb$_{18}$Co$_{78}$. The absorbed critical fluence calculated from ellipsometry measurements (see methods) increases linearly from 1.8 mJ/cm$^2$ to 2.5 mJ/cm$^2$ for a-Gd$_{22}$Co$_{78}$ and a-Gd$_4$Tb$_{18}$Co$_{78}$ respectively.

Time dynamics of HI-AOS of a-GdTbCo alloys

The temporal dynamics of the magnetization of the a-Gd$_{22-}\_x$Tb$_{78}$Co$_x$ films as they undergo HI-AOS was measured by time-resolved magneto optical Kerr effect (TR-MOKE) (see methods). An incident fluence of 6.9 mJ/cm$^2$ was chosen for all TR-MOKE experiments as it slightly exceeds the incident critical fluence of a-Gd$_4$Tb$_{18}$Co$_{78}$, the sample with the largest Tb content that exhibited HI-AOS. Fig. 2a shows the ultrafast magnetization dynamics of all samples studied measured with TR-MOKE. The magnetization reversal process follows a two-step behavior. In the first step a rapid initial drop in the magnetization occurs in which all films share a similar demagnetization process within the first picosecond post irradiation from the pump pulse. The second stage consists of remagnetization in the opposite direction as the system cools down, except for Tb$_{22}$Co$_{78}$. a-Gd$_{22}$Co$_{78}$ exhibits the fastest remagnetization time and increasing the Tb content systematically slows down this rate. The remagnetization rate plateaus with 15% and 18% Tb samples exhibiting similar dynamics. Finally, a-Tb$_{22}$Co$_{78}$ only demagnetizes upon irradiation and then recovers its magnetization along its initial direction upon cooling. It is possible that Tb$_{22}$Co$_{78}$ exhibits a transient switching in the first few ps following irradiation, similar to the behavior reported by Alebrand et al., and modeled by Moreno et al., suggesting that switching could occur at a higher fluence. However, utilizing higher fluences led to irreversible damage of the sample as the laser ablated or burned the sample surface. By 200 ps all samples had remagnetized to about 80% of the saturation value.

Fig. 2b is a close up of the experimental data of Fig. 2a, and it shows, after the initial fast demagnetization step, a slight bump in the magnetization that deviates from exponential decay behavior as evidenced in a-Gd$_{22}$Co$_{78}$, a-Gd$_{18}$Tb$_4$Co$_{78}$ and a-Gd$_{14}$Tb$_8$Co$_{78}$. The duration of this bump increases with increasing Tb, but for the higher Tb alloys the bump is more linear in character before resuming exponential decay characteristics.

Simulation of HI-AOS in a-GdTbCo alloys

Atomic spin dynamics simulations using the VAMPIRE software package combined with a two-temperature model (2TM) details discussed in suppl. matls.) were performed to simulate the experimental magnetization dynamics. As shown in Fig. 2c the simulation is in excellent agreement with the experiments in reproducing the characteristic behavior of similar demagnetization dynamics followed by increasingly slow remagnetization times with increasing Tb content. The bump in the magnetization following the initial demagnetization step is also reproduced in simulation, and exhibits a more linear character with increasing Tb as seen experimentally. The discrepancy in the time scales between experiment and simulation is due to both the small size of the simulated system not allowing for domain dynamics to be taken in consideration, and also to heat dissipation effects. We note here that the simulations were able to reproduce the experimental
FIG. 2. a) Time-resolved magnetization dynamics of α-
Gd$_{22-x}$Tb$_x$Co$_{78}$ thin films measured with a laser fluence of 6.9
mJ/cm$^2$. The initial rapid drop in magnetization is similar across all
samples, but upon entering the remagnetization regime different rates
are observed. b) Close up of the experimental data showing the bump
in the magnetization following the initial demagnetization step. c) Simulated
magnetization dynamics of Gd$_{22-x}$Tb$_x$Co$_{78}$ after laser irradiation obtained with a two-temperature model ne-
glecting spin-lattice coupling as described in the text.

Effect of annealing on switching dynamics

The effect of annealing to test the influence of anisotropy and damping on the dynamics is shown in Fig. 3a. Annealing α-Gd$_{10}$Tb$_{12}$Co$_{78}$ at 300 °C for one hour results in a significant reduction in coercivity and anisotropy (from 4.6 × 10$^5$ erg/cm$^3$ to 2.5 × 10$^5$ erg/cm$^3$) while maintaining the composition and $M_S$ constant as seen in Fig 3b. Further annealing the sample at 350 °C removed the PMA. The fact that $M_S$ was unchanged by annealing strongly indicates that inhomogeneities such as phase segregation have not occurred. A slower remagnetization time is observed upon annealing in the magnetization dynamics shown in Fig. 3b, similar to those in the 15% and 18% Tb samples. Atomistic simulations of the magnetization dynamics of this sample as a function of Gd damping are shown in Fig. 3c. It can be seen that increasing the damping on the Gd sites explains the slower remagnetization time, suggesting that the experimentally annealed sample has an increased damping, in addition to the reduced anisotropy. Indeed, work from Malinowski et al. showed that introducing local variations of the anisotropy in amorphous CoFeB leads to an increase in the damping parameter. Since the origin of anisotropy in RE-TM alloys is due to a combination of pair-ordering along the growth direction and the single ion anisotropy of Tb$^{2+}$ annealing leads to a structural relaxation of the pair-ordering that introduces local anisotropy variations which in turn lead to higher damping and thus the slower remagnetization time observed. As observed from Fig. 2, the samples with higher anisotropy (larger Tb at.%) show slower switching dynamics. From our annealing studies, we observe that the film with lower anisotropy exhibits slower switching. These observations lead us to conclude that it is the damping of the system, and not the anisotropy, that is the significant contributor to the ability to exhibit HI-AOS. Simulations with varying anisotropy back this conclusion.

DISCUSSION

Fig. 4 shows the simulation results of critical fluence as a function of Tb concentration and varying Gd damping. The...
FIG. 3. a) Magnetization dynamics of $a$-Gd$_{10}$Tb$_{12}$Co$_{78}$ in the as-grown state (blue curve) and after annealing (red curve) at 300 °C for 1 hour. Annealing leads to a slower remagnetization time. b) Magnetization loops in the out-of-plane orientation depicting how annealing reduces the coercivity and anisotropy while keeping $M_S$ intact. c) Simulated time-resolved magnetization dynamics of $a$-Gd$_{10}$Tb$_{12}$Co$_{78}$ as a function of increasing Gd damping. Increasing the damping leads to a slower remagnetization time, indicating that annealing leads to a higher damping value.

critical fluence for switching in the simulation is determined by the transition from non-deterministic to deterministic thermally induced switching. It shows that increasing the Tb content increases the critical fluence as observed experimentally, regardless of the specific Gd damping value used. It also shows, that for a given concentration, increasing the damping on the Gd site increases the critical fluence. The simulations strongly indicate that the relative element specific damping of the rare earth site compared to the cobalt site is the key factor that influences the critical fluence required switching and the speed of remagnetization. As verified in the model for the annealed sample, at a fixed composition the key parameter that leads to slower remagnetization is the increase in damping at the Gd site. In simulation the damping constant is a phenomenological parameter that combines a host of diverse effects whose physical origins are not often well defined. Increasing the Tb composition leads to a greater spin-orbit interaction in the system, which is considered the intrinsic source of damping and is proportional to $\xi^2/W$, where $\xi$ is the spin orbital coupling energy and $W$ is the d-band width. Thus as the system becomes Tb-rich it experiences increased spin-orbit coupling which increases damping and thus leads to the dynamics observed in Fig. 2. For the dynamics observed in the annealed sample, the spin-orbit coupling may not have changed drastically, but the anisotropy of the coupling likely did due to the structural relaxation undergone after annealing, thereby increasing the macroscopic damping. The simulation does not directly account for spin-orbit coupling, but it indirectly simulates the effects of stronger coupling via increases in the damping parameter. As mentioned before the deterministic switching is independent of the anisotropy and therefore the main contribution of spin-orbit coupling for all optical switching is the damping. Therefore although the simulations reveal the critical role of damping in modifying the ultrafast magnetization dynamics, it is entirely possible that the underlying physical mechanism is rooted in the spin-orbit interaction.

FIG. 4. Simulated critical fluence as a function of Tb concentration for different damping values of Gd. The Tb and Co damping parameters are constant (0.05). The critical fluence increases as the damping of Gd increases, indicating that lowering the damping of Gd reduces the threshold switching condition of Gd-Tb-Co alloys below the laser ablation limit.
CONCLUSION

In conclusion, we have shown that \(a\)-Gd\(_{22-x}\)Tb\(_x\)Co\(_{78}\) thin films exhibit HI-AOS from \(x = 0\) to \(x = 18\), displaying a two-step reversal process with an identical fast demagnetization step and second slower remagnetization. The remagnetization time and the critical fluence increase as the Tb content increases, indicating that replacing Gd with Tb atoms hinders the switching process. Atomicistic simulations can explain the switching dynamics of our material system only after taking into account the individual element specific damping of each RE and TM sites, as opposed to a net damping of the whole system. The slower remagnetization dynamics and the increased critical fluence as the Tb atomic percentage increases are explained on the basis of increased damping on the RE site upon addition of Tb. Annealing of RE and TM sites, as opposed to a net damping of the whole film led to a slowing of the remagnetization rate, as which as verified by simulations indicates that damping is responsible for modifying the ultrafast magnetization dynamics. The experimental inability to observe HI-AOS in \(a\)-TbCo is due to the high critical fluence that under the present conditions is inaccessible without ablating the film. This suggests that better management of the laser thermal load may lead to HI-AOS beyond Gd-TM alloys. Our results indicate that the engineering of element specific damping of RE-TM systems will be crucial in our endeavor to uncover material systems exhibiting HI-AOS with favorable properties (such as high anisotropy and large magnetization) for applications in ultrafast spintronic devices.

METHODS

Sample growth and characterization

Samples were deposited at room temperature using a magnetron sputtering system by co-depositing from separate Tb, Gd and Co targets at an Ar pressure of 1 mtorr and a background pressure of \(6 \times 10^{-8}\) torr (see methods for sample growth and characterization details). Layer thicknesses were monitored with a quartz mass balance during growth and then confirmed via X-Ray reflectivity analysis. Rutherford backscattering spectrometry was used to measure the composition. Energy dispersive spectroscopy images taken with a scanning transmission electron microscope found no evidence of inhomogeneities at the 10 nm scale as reported in Gd-Fe-Cd\(^{[19]}\) (See Suppl. Matls.). The magnetization was measured with a SQUID magnetometer and found the compensation temperature (\(T_M\)) of all samples to be near 400 K due to the fixed 22 at.% RE content, allowing to maintain nearly constant values of saturation magnetization (\(\sim 100\) emu/cc) and \(T_M\) while varying the Gd/Tb ratio. At room temperature, the magnetization of all the films studied is RE-dominant. A vibrating sample magnetometer found the Curie temperature to lie above 600 K. This lower limit is given because as the temperature increases, the PMA decreases and is eliminated in the 620 - 670 K range\(^{[20]}\) and at these high temperatures the films are irreversibly modified by the nucleation of crystallites.

MOKE microscopy of single-shot switching events

The samples are irradiated with 100 fs pulses from a regeneratively amplified Ti-Sapphire laser, with a central wavelength of 810 nm and a bandwidth of 50 nm. The laser beam is p-polarized (electric field in the plane of incidence), and is focused to an elliptical spot with a FWHM of \(\sim (110 \times 80) \; \mu\text{m}^2\), incident at an angle of 50° with respect to the sample normal. Magneto Optical Kerr Effect (MOKE) imaging is done with a 630 nm LED. The images from the MOKE microscope are then processed to enhance the contrast arising from changes in magnetization (shown in Fig 1a by the light and dark grey regions).

Absorbed fluence calculation

Ellipsometry measurements yielded a complex refractive index of \(3.5 + 4.2i\) for all samples as extracted from multilayer absorption calculations using the transfer matrix method\(^{[20]}\).

Time-Resolved MOKE

Time-resolved MOKE (TR-MOKE) measurements were performed by splitting the laser beam into pump and probe beams and focusing them at the same spot on the sample. The pump beam is as described in the section above for the single shot experiments and triggers the HI-AOS and demagnetization processes. The probe is incident normal to the sample surface and its polarization is modulated at 50 kHz by a photo-elastic modulator (PEM). The arrival time of the probe at the sample relative to the pump is varied by passing the pump beam through a linear delay stage. This enables the detection the temporal dynamics of the magnetization of the \(a\)-Gd\(_{22-x}\)Tb\(_x\)Co\(_{78}\) films as they undergo HI-AOS by sending the photodiode signal from the reflected pump into a lock-in amplifier referenced at the PEM frequency. An incident pump fluence of 6.9 mJ/cm\(^2\) was chosen for all TR-MOKE experiments as it slightly exceeds the incident critical fluence of \(a\)-Gd\(_4\)Tb\(_{18}\)Co\(_{78}\), the sample with the largest Tb content that exhibited HI-AOS.

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AUTHOR CONTRIBUTIONS

A.C., A.P., A.E-G., F.H., and J.B. conceived and planned the experiments. A.C. and C.S. grew and characterized the samples. Laser measurements were performed by A.P. and A.E.-G. Simulations were performed by S.R. on the VAMPIRE software package developed by T.O., R.W.C. and A.E.-G. A.C., A.P., A.E-G., F.H., and J.B. conceived and planned all-optical magnetization switching mechanisms using femtosecond laser pulses; Physical Review B 96 (2017), 10.1103/PhysRevB.96.220411

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