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ABSTRACT

Repeated uniform switching of the magnetization of thin films of ferrimagnetic amorphous $Gd_x(FeCo)_{1-x}$ in response to single fast laser pulses is well established. Here, we report unusual toggle switching in thin films of sperimagnetic amorphous Dy_xCo_{1-x} and Tb_xCo_{1-x} with $x \approx 0.25$ irradiated with single 200 fs pulses of 800 nm laser light. The samples have strong local random anisotropy due to the non-S state rare earth. The compensation temperature of the films is $\leq$180 K, and their Curie temperature is $\approx$500 K. They are largely switched by the first pulse, and subsequent pulses lead to partial re-switching of a decreasing amount of the irradiated area, with a granular structure of sub-micrometer regions of switched and unswitched material. Individual switched regions about 700 nm in size are observed around the edge of the irradiated spots where the fluence is at the threshold for switching. Results are discussed in terms of a random anisotropy model where the ratio of local anisotropy to exchange is temperature dependent.

Manipulation of magnetization in thin films without applying a magnetic field is of great interest in spin electronics. Conventional methods of switching the magnetization by using an external directional stimulus, such as a pulsed magnetic field or a spin-polarized current, are limited to timescales of about 100 ps.[1] Single ultrafast laser pulses with durations of an order of 100 fs were originally shown by Beaurepaire *et al.* to demagnetize a nickel film in about a picosecond.[2] Subsequently, single-pulse all-optical thermal switching (SP-AOS) of the magnetization on a picosecond timescale was established by Ostler *et al.* in ferrimagnetic amorphous GdFeCo with an atomic Gd concentration $x \approx 25\%$,[3] and its observation was extended to other Gd-based thin film systems.[4,5] The notation "a-" will be used to denote an amorphous alloy. The first non-Gd based ferrimagnetic film to exhibit the effect was crystalline Mn_2Ru_xGa,[6,7] where Banerjee *et al.* have shown that re-switching is possible after 10 ps.[8]

The study of Radu *et al.* in 2011 regarding the dynamics of the Gd and Fe subnetworks during spin reversal in their a-GdFeCo films after a 50 fs laser pulse found that an unexpected transient parallel alignment of the Gd and Fe/Co moments preceded switching.[9] The same group confirmed that SP-AOS was essentially a thermal effect.[3] The magnetization could be repeatedly toggled back and forth between two orientations, and the effect was independent of the helicity of the light.

A general thermodynamical framework for this ultrafast laser-induced spin dynamics in multisublattice magnets was proposed by Mentink *et al.*[10] A temperature-dominated regime yields distinct sublattice demagnetizing dynamics because of the different magnetic moments, with the Fe/Co sublattice reaching zero first.[9] On the picosecond timescale, interatomic exchange of angular momentum (exchange relaxation) is dominant and brings about the ferromagnetic-like state by transferring angular momentum from the Gd to the fully demagnetized Fe. The magnetization of the Fe then increases in a direction opposite to the original one as it cools, while the Gd moment is still decreasing, leading eventually to complete switching of the ferrimagnetic magnetization.

The phenomenon of helicity-dependent AOS requiring many short pulses has a quite different origin; it was discovered by Stanciu *et al.*[11] and it has been explored in a wide range of multilayers and alloy magnetic films.[12,13]

TABLE I. Characteristics of the samples. Magnetic values are at 296 K.

Nominal composition	Thickness (nm)	Capping layer	RMS roughness (nm)	T_{comp} (K)	M_s (kA/m)	M_s (μ_B/f.u.)	$\mu_0 H_c$ (mT)	Laser fluence threshold (mJ/cm^2)
$Tb_{0.25}Co_{0.75}$	20.0	Al_2O_3	0.34	20	240	0.41	44.3	7.0
$Dy_{0.25}Co_{0.75}$	10.3	SiO_2	0.36	180	175	0.29	35.0	2.6

Gd is a spherically symmetric ion, and a-GdFeCo is ferrimagnetic, because of the negative rare-earth (RE)–transition metal (TM) exchange interaction. However, for non-S-state amorphous RE–TM alloys such as a-TbCo or a-DyCo, the random local electrostatic fields acting on the non-spherical 4*f* charge distributions create random local anisotropy that influences the orientation of the RE moments in the magnetically ordered state. The dominant contribution of the RE to the net ferrimagnetic magnetization below the magnetic compensation point T_{comp} where the net RE and TM subnetwork moments cancel is much reduced in a sperimagnetic ground state where the Co subnetwork is essentially collinear but the Dy or Tb subnetwork moments are distributed at random in a cone whose axis is antiparallel to the Co[14] [see Fig. 3(b)].

After SP-AOS was discovered in a-GdFeCo thin films, much work was done to search for the effect in systems with a different RE. This met with little success. Films of a-TbCo[12,16,17] were found to exhibit multi-pulse helicity-dependent AOS at compositions around 25 at. %. Film thickness and the size of the laser spot relative to domain size were criteria for observing the effect.[16] SP-AOS is observed in series of a-$(Gd_{0.22-x}Tb_x)Co_{0.78}$ alloys for all except the x = 0.22 Tb end member.[18] Transient subpicosecond reversal has been seen in studies of the demagnetization dynamics of a-$Tb_{0.26}Co_{0.74}$.[19] Single circularly polarized 150 fs laser pulses have been found to create skyrmions as small as 150 nm in diameter in a-$Tb_{0.22}Co_{0.60}Fe_{0.9}$.[20] A study by Liu *et al.*[21] used two-wire gold nanoantennas placed on a-$Tb_{0.17}Fe_{0.72}Co_{0.11}$ thin films for near-field enhancement of the laser pulse to reversibly switch single 50 nm domains in the film. Furthermore, there is a recent report of SP-AOS in Tb/Co multilayer electrodes in a magnetic tunnel junction, but not of the amorphous alloy.[22]

Based on these results, we have examined a-Tb_xCo_{1-x} and a-Dy_xCo_{1-x} alloys with $x \approx 0.25$ for evidence of SP-AOS and signs of the influence of local random anisotropy, with a view to obtain a fuller understanding of the phenomenon.

All thin films were deposited by DC magnetron sputtering at room temperature on Si wafer substrates covered with 500 nm thermal oxide. The thin a-Dy_xCo_{1-x} layers were deposited in a Shamrock system with a base pressure of 2×10^{-8} Torr. The a-Tb_xCo_{1-x} thin films were grown using the ultra-high vacuum multichamber deposition and characterization tool *Trifolium Dubium*. They were co-sputtered from RE (Dy or Tb) and Co targets. Composition was controlled by changing the RE target plasma power while fixing the Co power. Samples were capped with a protective layer of either SiO_2 or Al_2O_3. A superconducting quantum interference device (SQUID) magnetometer was used to measure magnetization and hysteresis. Both films studied here exhibit perpendicular anisotropy. Hysteresis was also measured in an Evico Magnetics magneto-optical Kerr (MOKE) microscope and by anomalous Hall effect. Structural and magnetic information on two representative samples is summarized in Table I.

The x-ray diffraction measurements in Fig. 1 show only peaks associated with the silicon substrate, including a broad forbidden (002) peak due to stacking faults in the Si. There is perpendicular anisotropy and a compensation temperature in these films.[23] Fitting small angle x-ray reflection gives values for the film thickness and density. The RMS surface roughness for both films deduced from atomic force microscopy (AFM) is 0.34 nm.

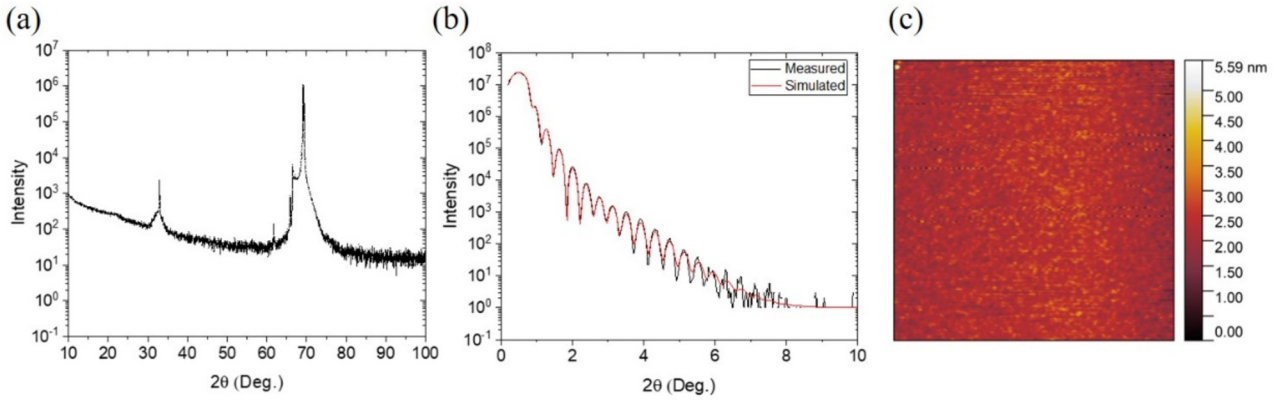

FIG. 1. (a) X-ray diffraction, (b) x-ray reflectivity, and (c) AFM characterization of the $Tb_{0.25}Co_{0.75}$ film. The area in (c) is $5 \times 5\ \mu m^2$.

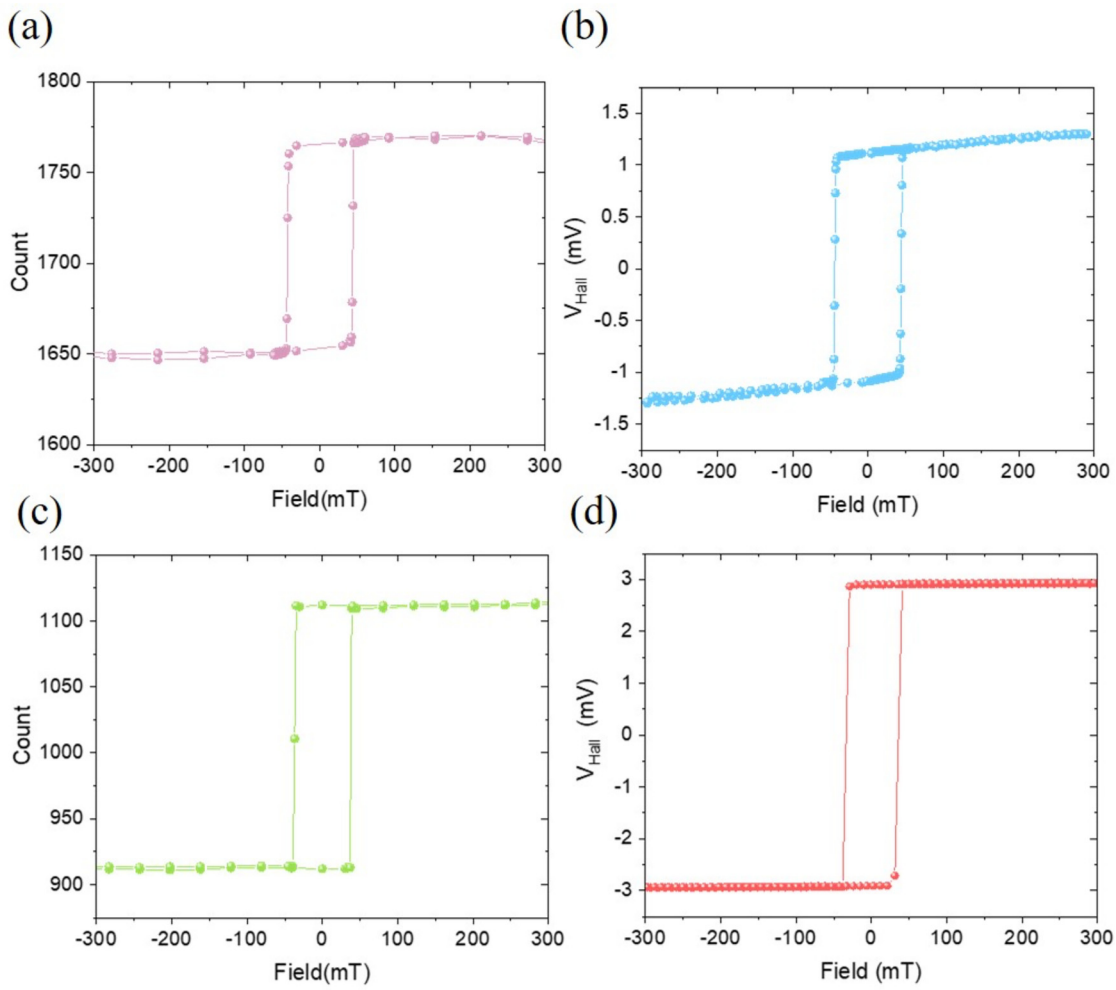

FIG. 2. Room-temperature hysteresis loops measured for a-$Tb_{0.25}Co_{0.75}$ by polar MOKE in blue light (a) and anomalous Hall effect (b) and for $Dy_{0.25}Co_{0.75}$ (c) and (d).

Some hysteresis loops of the films are presented in Fig. 2 where the data reflect the Co magnetization. The same coercivity is measured in the SQUID loops. The RE–TM coupling is antiferromagnetic, but the ferrimagnetism is modulated by random anisotropy on the RE that increases the net magnetization above T_{comp} and reduces it below T_{comp}. The magnetic ordering temperature deduced from extrapolation of the temperature dependence of the Dy sample is 500 K. The compensation temperature, where the net magnetization crosses zero and the coercivity diverges, is 180 ± 3 K for a-$Dy_{0.25}Co_{0.75}$. The Curie temperature of the Tb sample is expected to be similar, but T_{comp} is 20 ± 5 K, due in part to the lower Tb moment. A contribution to the magnetization of 1.5 μ_B per Co is deduced from measurements of a similarly prepared ferromagnetic a-$Y_{0.25}Co_{0.75}$ film, and the net moments of Dy and Tb at RT are deduced as 3.3 μ_B and 2.9 μ_B, respectively. They are much reduced from the atomic values of 10 μ_B and 9 μ_B by random anisotropy and weak RE–TM exchange.

The effect of irradiation by 1–10, 200 fs laser pulses was investigated *ex situ* using the MOKE microscope. The 800 nm pulses were generated by a Ti:sapphire laser amplifier operating in single-pulse mode. The pulse energy was controlled by rotating the half-waveplate (HWP), followed by the linear polarizer in the pulse beam path [Fig. 3(a)]. A parabolic mirror focused the beam with Gaussian intensity profile onto the sample surface down to a spot 90 μm in diameter, full width at half-maximum, calibrated using the Liu method.[24] Single pulse energy was determined by measuring the average beam power with the laser amplifier operating at 1 kHz. Prior to irradiation, the net magnetization M_{net} was saturated in a 0.5 T field directed outward. For both samples, the magnetization of the Co network is dominant at room temperature and initially in the outward direction as shown in Fig. 3(b). This state appears as the uniform gray area in the unirradiated films seen in Fig. 3(c). On irradiating with a single 800 nm pulse above a certain fluence threshold, an irregular black patch of switched magnetization appears at the center of image, where the net magnetization now points inward. Changing the laser pulse polarization to circular has no visible effect on the measured pattern. The threshold fluence is lower for the 10 nm thick $Dy_{0.25}Co_{0.75}$ than for 20 nm thick $Tb_{0.25}Co_{0.75}$ as seen in Table I. It increases with film thickness up to 40 nm. Switched areas after the first pulse, in Figs. 3–5, can be seen,

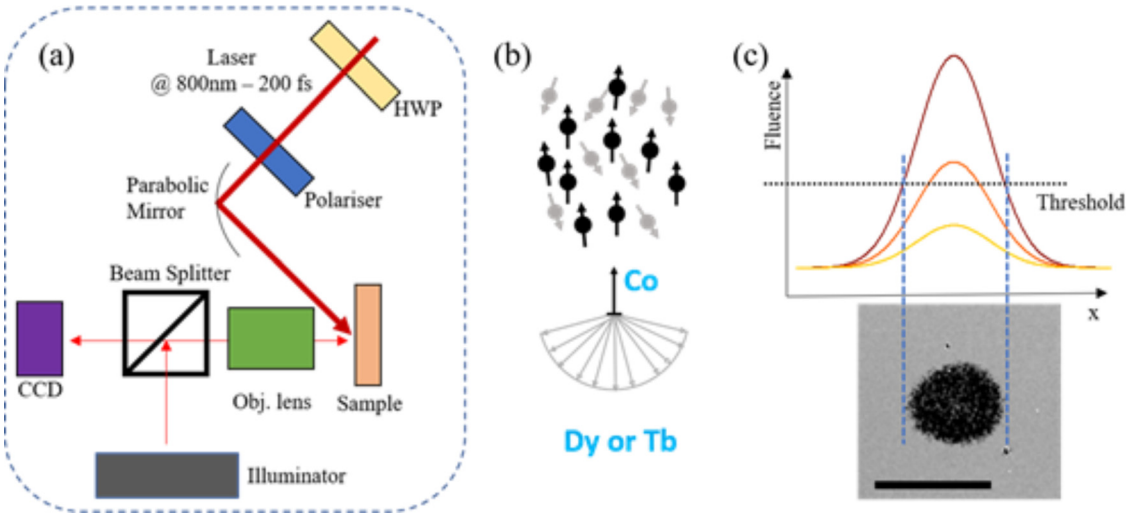

FIG. 3. (a) Experimental setup using a parabolic mirror to focus a single 800 nm laser pulse onto the sample surface, (b) sperimagnetic structure of a-DyCo below T_{comp}, and (c) Gaussian profile of the fluence on the sample. Above a certain threshold fluence, the magnetization of the $Dy_{0.25}Co_{0.75}$ sample after a single pulse adopts a mostly reversed, but magnetically non-uniform state. The scale bar is 50 μm.

especially in the diffuse border region, to be composed of regions $0.7 \pm 0.3\,\mu$m in size. This behavior is quite different to SP-AOS in similar conditions in a-GdFeCo or crystalline MnRuGa films, where no such structure has been detected[3,6] and the whole area irradiated above threshold is uniformly switched.

We systematically irradiated regions of the samples with sequences of one to ten pulses and examined them in the MOKE microscope. Data are shown in Fig. 4 for two different fluences and sequences of one to six pulses at each sample, separated by a 1 s delay. After an initial, nearly complete switch with the first pulse, the contrast between subsequent pulse pairs is reduced. The toggle switching was still evident after six pulses, but the change diminishes with successive shots.

Figure 5 presents a detailed analysis of higher-magnification (16 bit grayscale) images. The background is flattened away from the region of interest using a biquadratic polynomial. The normalization, based on the remnant state of the initially outwardly magnetized film, is accurate to within 5%. Histogram distributions of the local normalized data are constructed from the grayscale distributions within elliptical regions of interest that are fully centered within the irradiated spots. Figure 5(b) shows the distributions for the unperturbed film (0 shots) and after the first 1–3 shots; Fig. 5(c) shows the peak of the magnetization distribution after each sequential shot. Error bars signify the positive and negative half widths at half maximum of the distributions. The first pulse reverses 75% of the domains, the next one 45% and after ten pulses, only 10% is being reversed. The average size of the isolated domains is $0.7 \pm 0.3\,\mu$m, like that found in helicity dependent AOS of a-TbCo.[17] Figure 5(d) shows the switching of the a-$Dy_{0.25}Co_{0.75}$ sample with four different fluences. In each case, the outer annulus of the irradiated spot was analyzed.

The single-pulse all-optical toggle switching that we observe in Figs. 4 and 5 differs from that in a-GdFeCo and MnRuGa, where switched areas are uniform and well-defined. The nanoscale granularity of the switched areas in a-$Dy_{0.25}Co_{0.75}$ and a-$Tb_{0.25}Co_{0.75}$ is particularly evident around the borders of the spots, where fluence is close to threshold. Only the study by Liu *et al.* using nanoscale antennas has reported SP-AOS of a 50 nm single domain, smaller than those we find here.[21]

Analytical[15] and computational[25] investigations of the effect of random local anisotropy on a ferromagnet, carried out in the 1970s, provide some useful insight. The anisotropy was treated in the Harris–Plischke–Zuckermann (HPZ) model,[15] where the first term in Eq. (1) is the interatomic exchange and the second is a locally random "crystal field" interaction:

$$\mathcal{H}_{\mathrm{HPZ}} = -2\sum_{i>j}\mathcal{J}_{i,j}\boldsymbol{S}_i \cdot \boldsymbol{S}_j - \sum_i D_i(\boldsymbol{e}_i \cdot \boldsymbol{S}_i)^2. \qquad (1)$$

Here, D_i is the local uniaxial anisotropy parameter, $\mathbf{e_i}$ is the local anisotropy direction, $\boldsymbol{S}_i$ is the total angular momentum of an atom, and $\mathbf{e_i} \cdot \boldsymbol{S}_i$ is its z-component along the local anisotropy axis. The key quantity in the model is α, the ratio D/J of random anisotropy to exchange, which is independent of temperature in the one-sublattice random ferromagnet. The length scale over which the magnetization axis wanders is $L = (1/9\alpha^2)\pi^4 a$, where a is the average interatomic spacing.[14]

In the present case, the average distance between rare-earth ions deduced from the densities of the films is 0.5 nm. Taking L as the observed domain size, we find $\alpha \approx 0.1$ at RT, which corresponds to a weak pinning regime where the magnetic axis wanders over distances $L \gg a$. However, when applying the HPZ model to our amorphous ferrimagnets, α will increase with the decreasing temperature unlike in ferromagnets, where the ratio of anisotropy to exchange terms in Eq. (1) is temperature independent. The exchange interaction of the rare earth with the Co subnetwork can be represented by a molecular field, which varies little with temperature below $T/T_c = 0.7$, so the ratio of anisotropy to exchange varies as $\langle S^2\rangle/\langle S\rangle \simeq \langle S\rangle$, the thermal average of the rare-earth moment, which increases quickly around and below RT.

We take the established mechanism for SP-AOS in a-GdFeCo as a basis for the discussion of a-TbCo and a-$DyCo_{Co}$. A study of ultrafast spin dynamics in magnetic alloys containing Fe, Co, Ni, Gd, and Dy, by Radu *et al.*,[26] found that the change in demagnetization time with

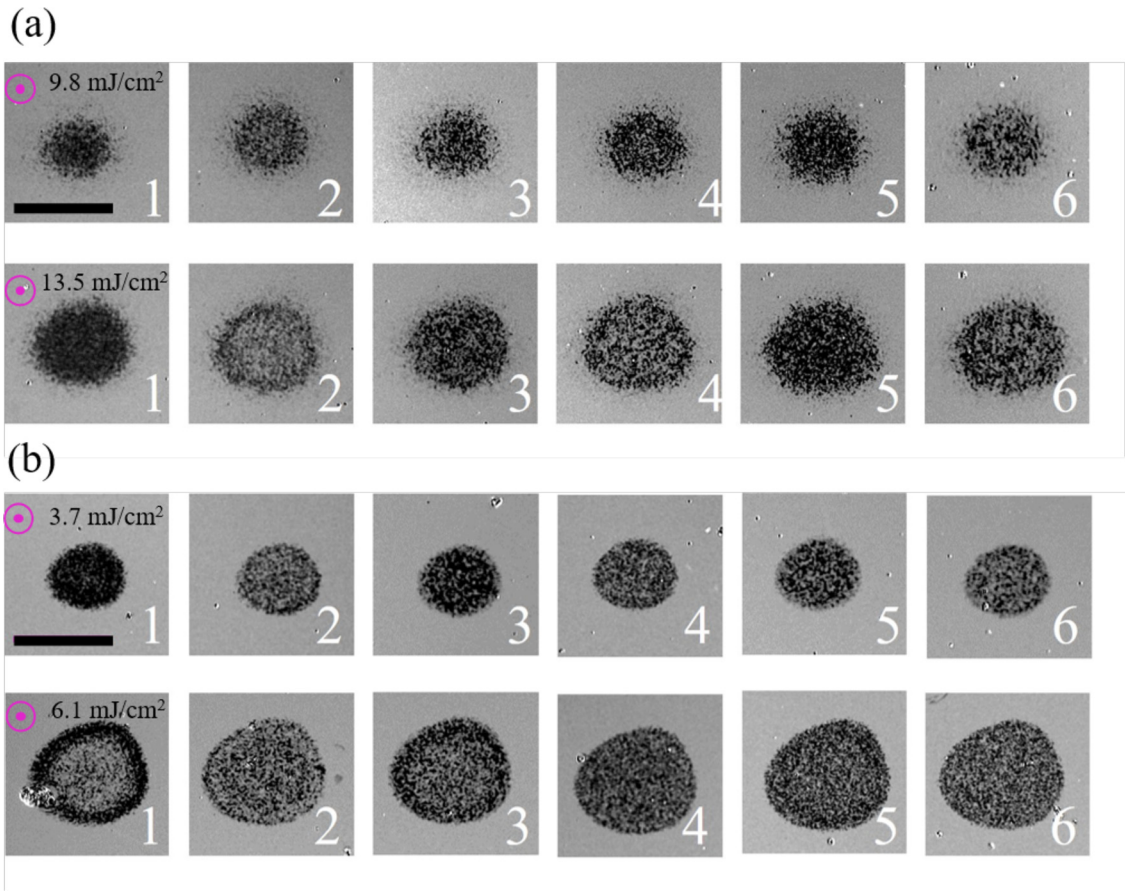

FIG. 4. MOKE images of films of (a) a-$Tb_{0.25}Co_{0.75}$ and (b) $Dy_{0.25}Co_{0.75}$ after a sequence of 200 fs laser pulses. The number of pulses is indicated on the figures. The fluences are noted at the beginning of each sequence. The scale bar is 50 μm.

atomic magnetic moment was 90 ± 10 fs/μ_B. The small atomic moment of Co is destroyed in less than 200 fs, but Dy or Tb will take much longer to demagnetize. Their lost angular momentum can be transferred to Co, once the Co atomic moment is re-established, and it re-orders parallel to the average rare earth moment. This produces, in the regions where the z-projection of the RE cone axis is sufficiently high, the transient ferromagnetic alignment that is crucial for SP-AOS. The restored Co subnetwork can now exert an exchange interaction on the disordered, but strongly correlated RE atomic moments, and as $\langle S \rangle$ increases on cooling, the rare-earth anisotropy imposes a new wandering axis on the switched ferrimagnetism.

Bulk perpendicular magnetic anisotropy is restored at some point in the re-magnetization process, which begs the questions of where the memory of the perpendicular anisotropy is stored, and when it becomes effective. Since we find that the magnetization of a-$Y_{0.25}Co_{0.75}$ is in-plane, we think that the positions of magnetic atoms in the amorphous structure create a two-ion dipolar anisotropy.[27] The stabilizing effect of this anisotropy builds up again after switching has occurred, and both RE and TM moments are restored.

In conclusion, the single-pulse all-optical partial switching, non-uniform on a length scale of ~700 nm that we observe in a-$Dy_{0.25}Co_{0.75}$ and a-$Tb_{0.25}Co_{0.75}$, is quite distinct from that in a-$Gd_x(FeCo)_{1-x}$ where the toggle switching is indefinitely repeatable and uniform. Random atomic-scale anisotropy on the rare-earth subnetwork of Dy and Tb alloys distinguishes them from their ferrimagnetic Gd counterpart.

The demagnetization rate of the high-moment atoms Dy and Tb is expected to be at least as slow as that of Gd, and both the formation of the transient aligned state and the re-magnetization process are influenced by the local random anisotropy, which increases faster than the exchange as temperature drops. The system at room temperature is in a weakly pinned region, consistent with an anisotropy direction that wanders over the ~700 nm switched regions.

Amorphous films containing REs with positive, negative, or zero electric quadrupole moment and positive or zero magnetic moment provide an opportunity to disentangle the factors that influence the de- and re-magnetization processes of these interesting magnetic materials. Extending the scope of picosecond timescale investigations from local magnetization to magnetic anisotropy on both local and mesoscopic scales will deepen our understanding of single-pulse all-optical switching in systems with strongly correlated 4f moments.

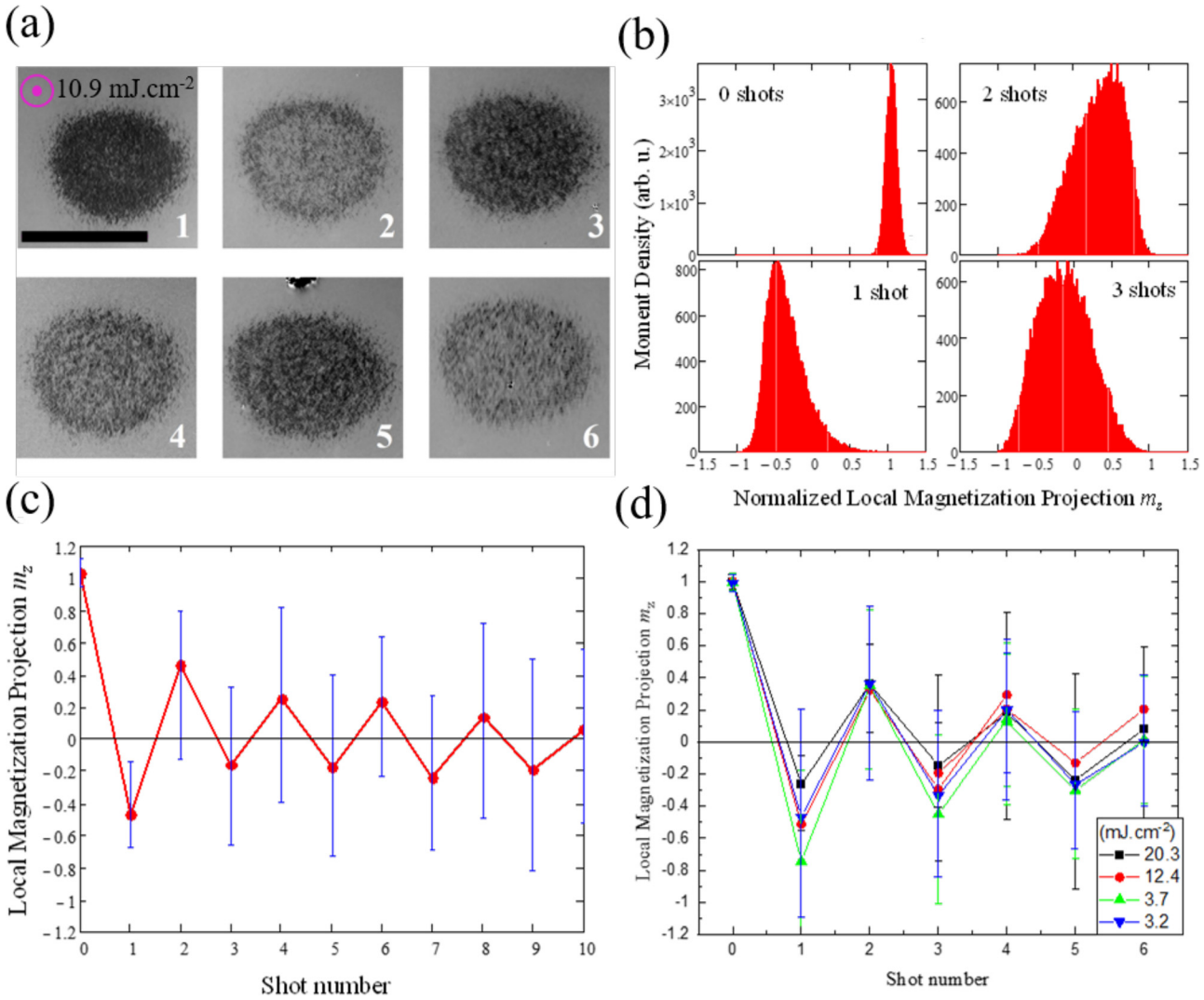

FIG. 5. (a) MOKE images of a-$Tb_{0.25}Co_{0.75}$ films obtained after a sequence of 200 fs laser pulses. The scale bar is 50 μm. (b) The domain size distribution of the a-$Tb_{0.25}Co_{0.75}$magnetized film, and after 0–3 pulses (c) The normalized fractions of outwardly or inwardly magnetized domains of a-$Tb_{0.25}Co_{0.75}$ after 0–10 pulses. (d) Switching measured for a-$Dy_{0.25}Co_{0.75}$ films irradiated with different laser fluences.

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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 available within the article.

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