












Magnetic and all-optical switching properties of amorphous Tb_xCo_{100-x} alloys

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Amorphous Tb_xCo_{100-x} magnetic alloys exhibit a list of intriguing properties, such as perpendicular magnetic anisotropy, high magneto-optical activity, and magnetization switching using ultrashort optical pulses. Varying the Tb:Co ratio in these alloys allows for tuning properties such as the saturation magnetic moment, coercive field, and the performance of light-induced magnetization switching. In this paper, we investigate the magnetic, optical, and magneto-optical properties of various Tb_xCo_{100-x} thin-film alloy compositions. We report on the effect the choice of different seeding layers has on the structural and magnetic properties of Tb_xCo_{100-x} layers. We also demonstrate that for a range of alloys, deposited on fused silica substrates, with Tb content of 24–30 at. %, helicity-dependent all-optical switching of magnetization can be achieved, albeit in a multishot framework. We explain this property to arise from the helicity-dependent laser-induced magnetization on the Co sublattice due to the inverse Faraday effect. Our paper provides an insight into material aspects for future potential hybrid magnetoplasmonic TbCo-based architectures.

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I. INTRODUCTION

The development of magnetic memory storage devices was accelerated by advancements in nanofabrication and material preparation methods. However, for dense and fast magnetic memory devices, optimal materials and nanostructures are essential. Rare-earth transition-metal (RE-TM) alloys have become a material class of interest after the first observation of all-optical switching (AOS) of their magnetization, using ultrashort laser pulses, as in the case of ferrimagnetic GdFeCo alloys [1,2]. Many studies have been carried out in the area of AOS, ranging from its observation in different classes of magnetic materials [3], such as ferrimagnetic alloys [4,5], compensated ferrimagnets [6], multilayers [7], and even ferromagnetic films [8,9] and ferro- and ferrimagnetic multilayers [10–13], e.g., Pt-sandwiched Co films [14], and Gd/Co bilayers [15], to the exploration of the parameter space for the observation of AOS. The latter includes mate-

rial compositions [16,17] and laser pulse parameters, such as fluence [18], helicity [19,20], and pulse duration [21]. A range of both experimental and theoretical works were performed to explain the mechanism of AOS: ultrafast heating [22,23], angular momentum transfer between sublattices [24] comparing ultrafast heating and coherent effects [25], numerical studies investigating different spin-coupling strengths [26], mean-field models of relaxation dynamics in ferrimagnets [27], and *ab initio* calculations [28] combined with atomistic spin-dynamics modeling [9,29].

A single-shot switching behavior has been observed in amorphous GdFeCo [1], Gd/Co bilayers [15], and Co/Pt layers [14], but not in other materials, including other ferrimagnetic alloys. Instead, most of the investigated materials show helicity-dependent all-optical switching (HD-AOS) in response to a train of multiple circularly polarized laser pulses [8,9,12]. Though single-shot switching of magnetization as observed in GdFeCo [4] is highly desirable for the scheme of magnetic data writing, the low spin-orbit coupling of Gd leads to a lower magnetic anisotropy in this material, which may pose problems for long-term information retention as well as the downscaling of the memory-bit size. In this respect, the high spin-orbit coupling of Tb renders TbCo a promising material, exhibiting larger magneto-crystalline anisotropy than Gd-based alloys, which could be more relevant for HD-AOS spintronic-based applications, if optimized for its efficiency [30]. Recently, a single-shot helicity-independent all-optical switching was demonstrated in Tb/Co multilayers, coupled to CoFeB electrodes by Avilés-Felix

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et al. [7]. To the best of our knowledge, this is the first and so far the only observation of single-shot-induced magnetization switching in a Tb-Co system. However, no helicity dependence was demonstrated. Also, it is not yet clearly understood how similar or distinct the dynamics in multilayers and alloys are or why the multilayers show AOS. It has been theoretically predicted that $\text{Tb}_x\text{Co}_{100-x}$ could exhibit (due to laser heating) thermally induced magnetization switching over a wide range of atomic compositions, creating considerations for potential suitability for the preparation of building blocks of non-volatile memory elements [30]. TbCo alloys also exhibit a favorable perpendicular magnetic anisotropy (PMA) [31–33], which makes them attractive for densely packed magnetic memory elements. It was recently shown by Rowan-Robinson *et al.* [34] that even after a lithographic process to fabricate nanosized truncated $\text{Au/Tb}_{18}\text{Co}_{82}$ nanocones, the out-of-plane magnetic anisotropy is preserved in the magnetic tip of the nanocone. The Tb and Co sublattices interact antiferromagnetically but their magnetic moments are uncompensated, giving rise to a nonzero net magnetic moment. Starting from a TM-magnetic moment dominated condition at a given temperature, reduction of the Co content in the $\text{Tb}_x\text{Co}_{100-x}$ alloy increases the coercive field, μ_0H_c , and decreases the saturation moment until no net magnetic moment exists. This point is called the magnetization compensation point x_{comp} , at which the ferrimagnet resembles an antiferromagnet. Upon passing through the compensation point to a RE magnetic moment dominated composition region, the net magnetic moment of the alloy begins to increase again. Alternatively, an alloy with a given composition can become a compensated ferrimagnet upon adjusting the temperature to the corresponding magnetization compensation temperature T_{comp} [33].

In this paper, we present results of a study of the magnetic properties and HD-AOS behavior of amorphous RE-TM $\text{Tb}_x\text{Co}_{100-x}$ alloys. We investigate the effect of the atomic ratio of Tb:Co, the thickness of the films, and the underlying nonmagnetic buffer layers on the structural and magnetic properties of the alloys. We also explore the influence of the laser pulse characteristics, such as fluence, number of shots, and sweeping speed, on the AOS performance $\text{Tb}_x\text{Co}_{100-x}$ layer. We demonstrate an increased range of the compositions, namely, from 24 up to 30 at. % of Tb, for which HD-AOS switching occurs with a laser pulse duration of 90 fs and 240 fs, depending on the measurement (multishot or sweeping beam, respectively), which are, a broader range, and a smaller pulse width, than, e.g., reported by Alebrand *et al.* [11]. In the work by El Hadri *et al.* [17], the domain size with respect to the laser spot size was used as a criterion for the observation of AOS, however, in our paper we show that it is not the only criterion and other parameters, such as the Tb:Co ratio and laser parameters, that allow for the observation of HD-AOS irrespective of the domain size. The mechanism for HD-AOS in $\text{Tb}_x\text{Co}_{100-x}$ samples investigated here is not a single-shot helicity-independent mechanism like in GdFeCo, but rather a cumulative multishot mechanism similar to that observed in FePt and Co/Pt multilayers [8,12,20]. To explain the origin of the HD-AOS, we further use *ab initio* calculations to compute the local magnetization that can be coherently induced by circularly polarized light through the inverse Faraday effect (IFE) [35]. Lastly, an aim of this paper is to establish a

suitable parameter space for the observation of HD-AOS in $\text{Tb}_x\text{Co}_{100-x}$ alloys, using a minimal number of laser shots.

II. MATERIALS AND METHODS

Amorphous $\text{Tb}_x\text{Co}_{100-x}$ alloys were prepared in an ultrahigh vacuum DC magnetron cosputtering system from elemental Tb and Co targets (99.99% purity) and using Ar as a sputtering gas. The sample holder was rotating during deposition to ensure film thickness and composition uniformity. The following stacking structures were fabricated: substrate/spacer/ $\text{Tb}_x\text{Co}_{100-x}$ /capping, where substrate was fused silica, spacer was either $\text{Al}_{80}\text{Zr}_{20}$, Al_2O_3 , or $\text{Au/Al}_2\text{O}_3$, or no spacer was used at all, and capping layer was $\text{Al}_{80}\text{Zr}_{20}$, Al_2O_3 for respective spacer layers, or Al_2O_3 capping was used for $\text{Au/Al}_2\text{O}_3$ spacers and for the samples without the spacer layers. TbCo films were then characterized by Rutherford backscattering spectrometry (RBS) and particle-induced x-ray emission (PIXE), and by x-ray scattering techniques to be able to probe the conditions for AOS observation for a well-defined composition and atomic structure of the fabricated $\text{Tb}_x\text{Co}_{100-x}$ layers. The elemental composition of the samples was investigated by ion beam analysis at the Tandem Laboratory of Uppsala University, using the 5-MV NEC-5SDH-2 tandem accelerator [36]. RBS and PIXE were employed using 2.0 MeV He^+ as a primary beam. X-ray reflectivity (XRR) measurements were performed to determine layer thickness as well as roughness. In the data presented in the figures, we refer to the nominal, i.e., as calibrated, thickness of the films. The XRR determined thickness was 2–4 nm smaller than the nominal one (see Supplemental Material [37]). The roughness of the layers as determined from fitting XRR curves using GENX [38], was around 5.9 Å for the samples deposited directly on the fused silica substrates. More values of the root-mean-square roughness of TbCo layers with varying Tb:Co ratios and buffer layers are provided in the Supplemental Material [37], Table I. The atomic structure of the films was investigated using grazing incidence x-ray diffraction (GIXRD) measurements to determine whether the layers are amorphous or crystalline. More details about the sample preparation and characterization can be found in the Supplemental Material [37] and in Refs. [39,40].

We further performed ellipsometry measurements to determine the optical constants, i.e., the refractive index n and the extinction coefficient k , within the visible and near-infrared range, for various concentrations of $\text{Tb}_x\text{Co}_{100-x}$. We measured 20 – 40 nm thick $\text{SiO}_2/\text{Tb}_x\text{Co}_{100-x}/\text{Al}_2\text{O}_3$ films with Tb content in the range of 18.4 – 30 at. % prepared onto fused silica substrates. For each sample, measurements were taken in the 400 – 1600 nm spectral range using different incidence angles in the range from 45° to 75° in the optical convention, i.e., from the sample normal. Finally, the fitting of the ellipsometric data and the extraction of the wavelength dependence of n and k was performed using the GENX software [38,41], specifically adapting it for the visible and near-infrared range and considering a stack layer model with thicknesses determined through XRR measurements and GENX fitting of the resulting reflectograms. More details about the n and k extraction are provided in the Supplemental Material [37].

Static magnetic and magneto-optical properties of the Tb_xCo_{100-x} films were analyzed employing magneto-optical Kerr effect (MOKE). Magnetization loops were measured using a polar MOKE (PMOKE) setup in reflection geometry with an incident light of 530 nm in wavelength. A maximum out-of-plane static magnetic field of 900 mT was accessible. The remanent magnetization state of Tb_xCo_{100-x} films was imaged using magneto-optical Kerr microscopy. We used a Kerr microscope with a white light source and applied the magnetic field perpendicular to the sample plane. To image the remanent magnetization state, the samples were first demagnetized in a time-dependent magnetic field of decaying amplitude. The obtained micrographs were then analyzed using a pair correlation function. Element-specific domain imaging combining x-ray photoemission electron microscopy (XPEEM) with the x-ray magnetic circular dichroism (XMCD) effect at the Co L_3 and the Tb M_5 edges [42] was performed at the surface/interface: Microscopy (SIM) beamline of the Swiss Light Source. XPEEM was further used to record x-ray absorption (XA) spectra for characterization of the chemical state.

To investigate HD-AOS of magnetization in the layer of Tb_xCo_{100-x} , static magneto-optical imaging was performed on $SiO_2/Tb_xCo_{100-x}/Al_2O_3$ samples (deposited directly on fused silica substrates and capped with Al_2O_3). The samples were illuminated at normal incidence using linearly polarized white light from an LED source. Upon transmitting through the sample, the light was collected by a 20 \times objective lens, passed through a crossed analyzer to obtain magneto-optical contrast, and eventually detected by a CCD camera. We studied the magnetization switching behavior as a function of sample composition and thickness for two types of ultrafast stimuli: (a) keeping the sample stationary and varying the number of laser pulses (shots) incident on the sample, and (b) sweeping the sample under a train of ultrashort laser pulses. To achieve HD-AOS, pump pulses were generated from a Ti:Sapphire amplified laser system with a 1 kHz repetition rate and a central wavelength of 800 nm. For the multishot measurements, the pulse width at the sample was 90 fs and incident at 15 $^\circ$ to the sample normal. For the sweeping beam measurements, the pump had a pulse width of about 240 fs at the sample position and was incident at 10 $^\circ$ to the sample normal. A translation stage was used to control the sweep speed for the sweeping beam measurements.

To investigate the origin of the AOS we performed *ab initio* calculations of the IFE [35] that quantifies the amount of magnetization induced in the material by circularly polarized laser light. To this end, we used the nonlinear response theory formulation [28,43], which gives the induced magnetization δM^{IFE} per simulation-cell volume as

$$\delta M^{IFE} = K^{IFE}(\omega)I/c, \quad (1)$$

where $K^{IFE}(\omega)$ is the frequency-dependent IFE constant, I is the intensity of the laser, and c is the velocity of light. The IFE constant was computed separately for the Tb and Co atoms, assuming here the ordered $TbCo_2$ Laves compound as model system.

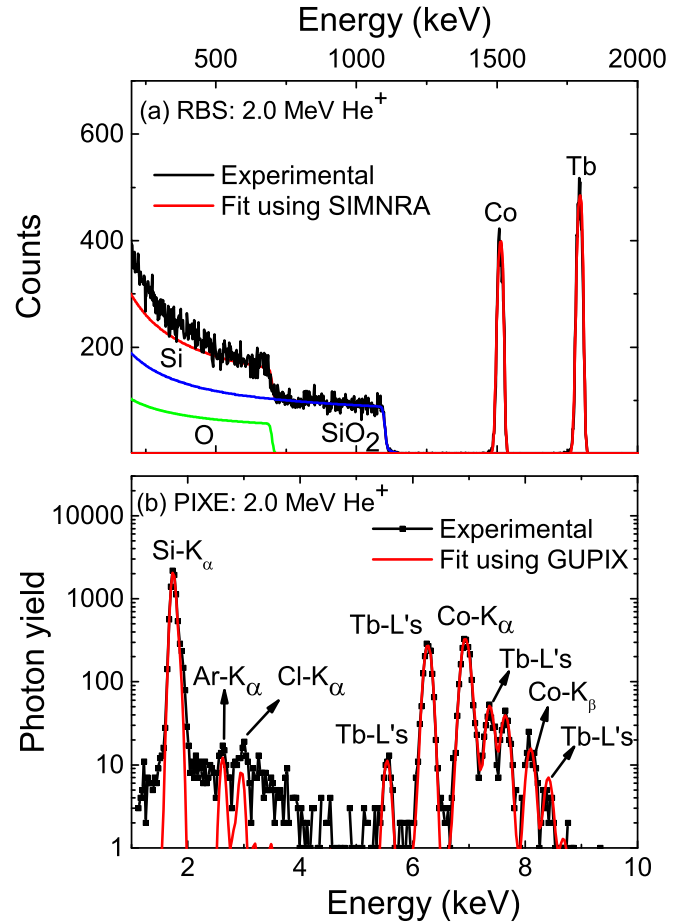


FIG. 1. (a) Experimental RBS spectrum recorded for the 2 MeV He^+ primary ions scattered from the $Tb_{18}Co_{82}$ sample prepared on a fused silica substrate (black solid line). The red solid line represents the best fit provided by the SIMNRA code [44]. The green and blue lines represent the contributions from the O and Si, respectively, which arise from the fused silica substrate. (b) The PIXE spectrum (black line with symbols) from the same sample in (a), which is obtained simultaneously with the RBS spectra. The fit using the GUPIX code [45] is also shown for comparison (red solid line).

III. RESULTS

A. Elemental and structural characterization

A typical RBS spectrum from $Tb_{18}Co_{82}$ sample studied in this paper is shown in Fig. 1(a). The statistical uncertainty of the Tb:Co ratio was found to be $\approx 2\%$, whereas systematic uncertainties were estimated to be lower than 1%. The elemental composition was determined for samples deposited under different deposition conditions to obtain the desired Tb:Co ratios. In Fig. 1(b), the PIXE spectrum of the same sample is shown. As can be seen in the PIXE fit, trace contaminations of Cl and Ar were found in the sample (≈ 0.2 at. %), while no evidence of heavy trace elements ($Z > 11$) was detectable in the film (quantification limit > 0.1 at. % for the present measurements). The Ar trace contamination found in the films was incorporated during the sample preparation since Ar was used as sputtering gas.

The structure of the fabricated Tb_xCo_{100-x} films prepared on fused silica substrates, without any buffer layer as well as

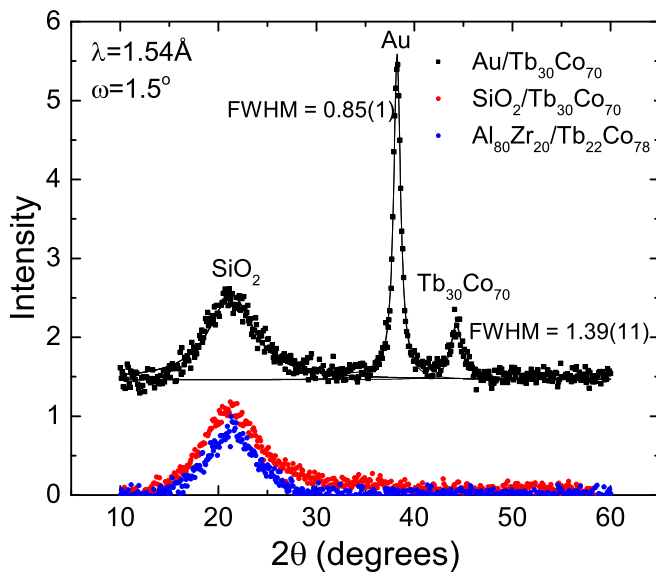


FIG. 2. Diffractograms of a $\text{Tb}_{30}\text{Co}_{70}$ sample deposited onto a fused silica substrate (red points) and onto a Au buffer layer (black points), both capped with Al_2O_3 , as well as of a sample deposited onto and capped with $\text{Al}_{80}\text{Zr}_{20}$ (blue points). Diffractograms were deconvoluted into Lorentzians, given by the solid black lines, and the intensity was normalized to the SiO_2 peak. The data of $\text{Au}/\text{Tb}_{30}\text{Co}_{70}$ sample (black measurement points) have been shifted upward for clarity.

with Au or $\text{Al}_{80}\text{Zr}_{20}$ buffer layers, was investigated employing GIXRD measurements. The $\text{Al}_{80}\text{Zr}_{20}$ buffer layers were used to ensure amorphicity of the $\text{Tb}_x\text{Co}_{100-x}$ films, as previously reported by Frisk *et al.* [32]. The Au buffer layers were chosen in this paper to investigate influence on the structure and thus possibility of directly combining $\text{Tb}_x\text{Co}_{100-x}$ alloys with typical plasmonic materials such as Au, a prerequisite for the fabrication of future $\text{Tb}_x\text{Co}_{100-x}$ -based magnetoplasmonic architectures. A typical GIXRD diffractogram for two $\text{Tb}_{30}\text{Co}_{70}$ films, one prepared on a fused silica substrate and the other on a Au buffer layer, and for a $\text{Tb}_{22}\text{Co}_{78}$ film, prepared on an $\text{Al}_{80}\text{Zr}_{20}$ buffer layer, are shown in Fig. 2. The films, prepared directly onto a fused silica substrate and on the $\text{Al}_{80}\text{Zr}_{20}$ buffer layer, exhibit a single, low-intensity broad peak at around $2\theta = 22^\circ$ that can be associated with amorphous SiO_2 (red symbols), while three peaks can be observed for the film prepared onto a 20 nm thick Au buffer layer (black symbols). The first peak, at $2\theta = 22^\circ$, similar to the previous case, corresponds to the substrate; the second, narrow high-intensity peak around $2\theta = 38^\circ$ arises due to the Bragg reflection from Au (111) planes, while the third, narrow low-intensity peak corresponds to the $\text{Tb}_{30}\text{Co}_{70}$ film.

In the work by Frisk *et al.* [32], it was shown that the amorphicity of $\text{Tb}_x\text{Co}_{100-x}$ films depends on the Tb content in the film and the crystallization onset is at around 80 at. % of Tb. This comparison shows that films deposited onto amorphous substrates, such as fused silica or Al_2O_3 , grow in an amorphous fashion, while deposition onto the polycrystalline buffer layers such as Au leads to a probable formation of nanocrystallites at the interfaces, similar to what has been reported by Liebig *et al.* [46]. Crystallite

sizes, computed using Scherrer's equation, are 97.9 Å and 58.7 Å for Au and $\text{Tb}_{30}\text{Co}_{70}$ layers, respectively. To further confirm the amorphous structure of the $\text{Tb}_x\text{Co}_{100-x}$ films, we performed extended x-ray absorption fine structure (EXAFS) measurements of a $\text{Tb}_x\text{Co}_{100-x}$ film deposited on a fused silica substrate, which reveal no long-range order of atoms in our films. A spectrum recorded with x rays tuned for the Co edge is shown in Fig. 2 of the Supplemental Material [37].

B. Magnetic and chemical characterization

We investigated how the magnetic properties of $\text{Tb}_x\text{Co}_{100-x}$ films are affected by the different nonmagnetic buffer layers on which they are deposited and imaged the remanent magnetization state. The effect of various Tb buffer layers on the coercive field, μ_0H_c , of a 20-nm-thick $\text{Tb}_{18}\text{Co}_{82}$ film is illustrated in Supplemental Material [37], Fig. 3. The μ_0H_c is largest for samples prepared on an $\text{Al}_{80}\text{Zr}_{20}$ buffer layer (364 mT) while the films deposited on a 20-nm-thick Au buffer layer appear to have a reduced μ_0H_c (170 mT). This can be related to the structure of the $\text{Tb}_x\text{Co}_{100-x}$ films: $\text{Al}_{80}\text{Zr}_{20}$ is known to promote the amorphous growth of the films, while a polycrystalline Au buffer promotes nanocrystallite formation in TbCo films at the interface (see Fig. 2) [46]. However, an increase in μ_0H_c (to 280 mT) can be observed when a $\text{Tb}_{18}\text{Co}_{82}$ film is deposited onto a hybrid buffer of $\text{Au}/\text{Al}_2\text{O}_3$, so $\text{Tb}_{18}\text{Co}_{82}$ has an interface with amorphous Al_2O_3 . This demonstrates how the higher degree of amorphicity allows for increased coercivity in $\text{Tb}_x\text{Co}_{100-x}$ alloys. We would like to emphasize the impact of having a nonmagnetic buffer layer which modifies atomic structure of the deposited magnetic layer and, hence, its magnetic properties. This occurs without any interlayer exchange interactions between the buffer layer and the magnetically active layer, which could be used for magnetic property modification, as in the study by Tang *et al.* [47], where [Co/Ni] multilayers were used for tuning the x_{comp} of TbCo.

We further analyzed how the coercivity changes by varying composition and sample thickness. We varied the Tb content in the $\text{Tb}_x\text{Co}_{100-x}$ films of 20, 30, and 40 nm in thickness, deposited on untreated fused silica substrates and on a hybrid $\text{Au}(20\text{nm})/\text{Al}_2\text{O}_3$ buffer layers. The results are summarized in Fig. 3. For films grown on the untreated silica substrates, the μ_0H_c diverges at around 23 at. % Tb both from Co- and Tb-rich sides which is characteristic to ferrimagnetic alloys. This region, indicated by the red hashed area, corresponds to x_{comp} at room temperature. Similar values of x_{comp} at room temperature were reported in work by Alebrand *et al.* [11]. Grey regions at the lowest and highest Tb content represent ranges where the films of the same thickness exhibits an in-plane magnetic anisotropy (IPMA) rather than out-of-plane or PMA. Our investigations also show that reduction of the thickness of TbCo films to below 10 nm results in the emergence of IPMA at the expense of the desired PMA (see Supplemental Material [37]). Similar to observations of the buffer layer influence on μ_0H_c (see Fig. 3 in Supplemental Material [37]), the $\text{Tb}_x\text{Co}_{100-x}$ films prepared on hybrid $\text{Au}(20\text{ nm})/\text{Al}_2\text{O}_3$ buffer layers exhibit larger coercive fields than their counterparts prepared directly on fused silica substrates.

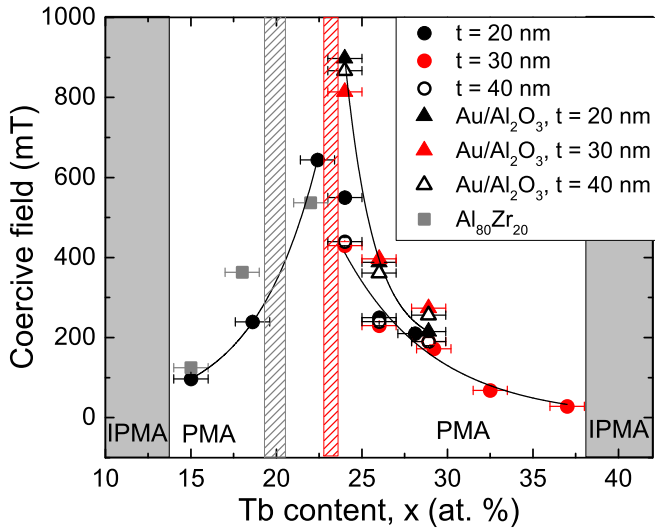


FIG. 3. Coercive field dependence on Tb_xCo_{100-x} composition for samples deposited on fused silica substrates (illustrated as circles), on fused silica/Au(20nm)/Al₂O₃(3 nm) buffer structures (triangles), and on Al₈₀Zr₂₀(3 nm) buffer layers (squares). Black, red, and empty symbols correspond to 20-, 30-, and 40-nm thickness of Tb_xCo_{100-x} layers, respectively. Correspondingly, IPMA and PMA denote regions, where the alloys exhibit an in-plane and perpendicular magnetic anisotropy. Red and grey hashed areas correspond to the x_{comp} for the sample series prepared directly on fused silica substrate and onto Al₈₀Zr₂₀(3 nm) buffer layers, respectively. Measurements were performed in polar MOKE configuration, with incident light of 530-nm wavelength.

In relation to the seed layer effect investigation, we also measured samples with 15, 18, and 22 at. % Tb prepared onto Al₈₀Zr₂₀ buffer layers. We observed that in contrast to films with 15 and 18 at. % Tb, the measured out-of-plane hysteresis loop of the sample with 22 at. % Tb appears to have changed the sign (see discussion in Supplemental Material [37]). This indicates that the compensation point for films prepared onto Al₈₀Zr₂₀ buffer layer is around 20 at. % Tb at room temperature, which is in agreement with the results of the previous study by Frisk *et al.* [32]. These results demonstrate the importance of an underlying buffer layer for the observed magnetic properties of the Tb_xCo_{100-x} films. In addition to investigations of RT x_{comp} , T_{comp} was determined by temperature-dependent polar MOKE measurements of the μ_0H_c of Tb_xCo_{100-x} films, prepared directly on the fused silica substrates. A Tb₁₈Co₈₂ film, deposited directly on a fused silica substrate, shows T_{comp} at around 170 K. For a film with 22 at. % Tb content, a slight increase of μ_0H_c is observed close to RT, which indicates that T_{comp} is slightly higher than 300 K, while divergence of μ_0H_c is not observed in the accessible temperature range (100 – 300 K) for Tb₂₄Co₇₆, which means that Tb_xCo_{100-x} films with $x > 22$ at. % Tb have a T_{comp} higher than 300 K. These observations emphasize the influence of a buffer layer on the magnetic properties of the ferrimagnetic Tb_xCo_{100-x} layers.

We imaged the magnetic domain structure of the 20-nm-thick Tb₂₄Co₇₆ film, deposited on fused silica substrates, using Kerr microscopy and XPEEM. Kerr microscopy image for this film shows a broad size distribution of the magnetic

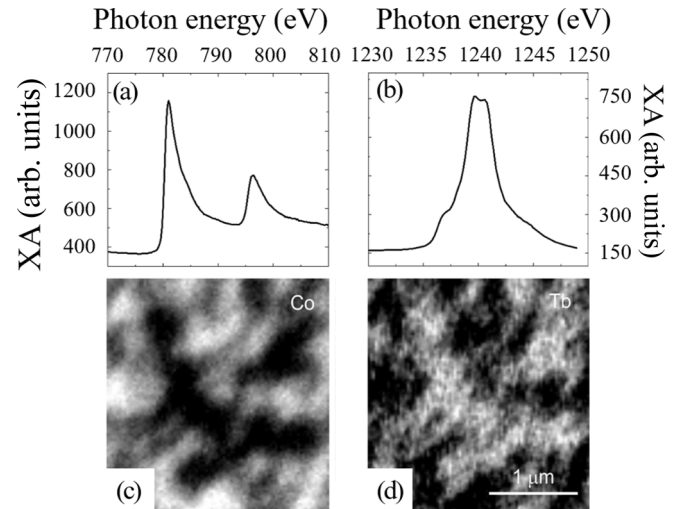


FIG. 4. X-ray absorption spectra measured at the (a) Co $L_{2,3}$ and (b) Tb M_5 absorption edges and magnetic contrast maps of a remanent magnetization state of a 20-nm-thick Tb₂₄Co₇₆ film obtained by combining XPEEM with XMCD at the (c) Co L_3 and the (d) Tb M_5 edges.

domains (see the inset of Fig. 7 in the Supplemental Material [37]). Figure 4(a) shows XA spectra recorded by means of XPEEM at the Co $L_{2,3}$ edges. The spectral shape corresponds to metallic cobalt without trace of oxidation [48]. Figure 4(b) shows the corresponding XA spectra at the Tb M_5 edge. Magnetic contrast maps recorded at the Co $L_{2,3}$ and the Tb M_5 edges are shown in Figs. 4(c) and 4(d), respectively, with domain sizes in the micrometer range. The contrast reversal between the domains visualized at the Co edge and the Tb edge directly confirms the ferrimagnetic coupling within the alloys.

C. Optical characterization

The wavelength dependence of the refractive index, n , and extinction coefficient, k , extracted from the ellipsometric data, is summarized in Fig. 5 for three alloy films with different Tb content within the 18–30 at. % range. For all samples, a monotonic increase with increasing wavelength is observed for both quantities. Similar qualitative behavior has also been recently reported for even higher concentrations of Tb [49]. It appears that there is no significant dependence on the Tb content for samples with a Tb content above the x_{comp} . However, for a sample with Tb content below compensation point, the n and k dispersion curves lie below the curves of samples with Tb content $x > x_{comp}$. Once again, the optical constants determined here have been targeted having in mind the potential of Tb_xCo_{100-x} films in magneto-plasmonic architectures (see, e.g., the work by Liu *et al.* [50], Rowan-Robinson *et al.* [34], and Cheng *et al.* [51]). For the proper electromagnetic design and simulations of these materials, knowledge of these optical constants is of paramount importance.

D. Magneto-optical imaging: All-optical helicity-dependent switching in the Tb_xCo_{100-x} layer

The conditions for observation of AOS were investigated in the Tb_xCo_{100-x} samples with varying x , deposited directly on

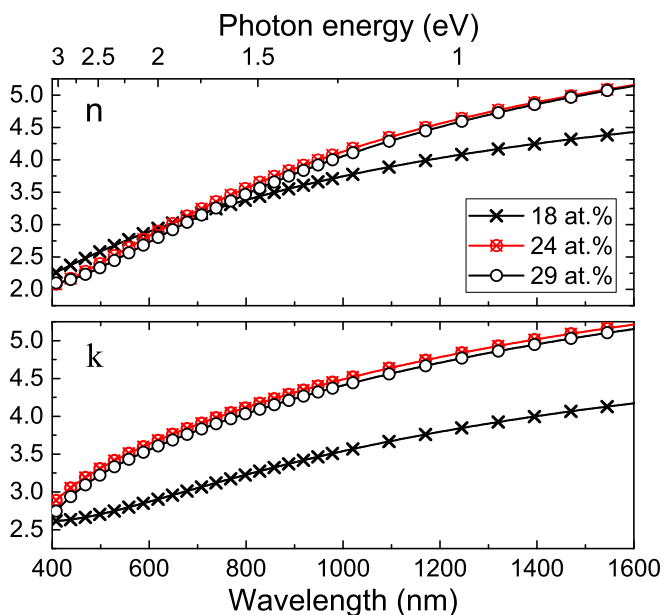


FIG. 5. Dispersion of the refractive index, n , and the extinction coefficient, k , for various Tb contents in a film, obtained from fitting ellipsometry data in the visible and near-infrared range. Layer structure and thickness were taken into account for ellipsometry data fits.

fused silica substrates and capped with Al_2O_3 3-nm layer *in situ*. The results of multishot and sweeping-beam experiments confirm the cumulative mechanism of HD-AOS in TbCo alloys, which is also confirmed by the theoretical calculations, discussed toward the end of this section. Single-shot helicity-independent switching of magnetization was not observed in any of the samples studied. However, a range of samples do exhibit a multishot HD-AOS under suitable laser pulse parameters, such as fluence and sweeping speed. The range of concentration and thickness, where HD-AOS was observed (for these samples), is plotted in the diagram of Fig. 6(a). Below 24 at. % Tb content, thermal demagnetization is always observed. This is shown in the left panel of Fig. 6(b), where both of the incident helicities, σ^+ and σ^- , induce a contrast change in two oppositely oriented magnetic domains to an intermediate gray, indicating that both helicities result in demagnetization.

Samples with Tb content ranging from 24 to 32 at. % show HD-AOS, as seen in the right panel of Fig. 6(b). For these samples, only one helicity can induce switching for a domain of a given orientation. The opposite helicity leaves the domain unaffected, showing a clear helicity dependence of domain writing. Further, for samples with $x > 32$ at. %, domain contrast cannot be observed with the polar Faraday imaging geometry, due to the shifting of easy out-of-plane magnetization axis toward the in-plane direction. Hence, it is unresolved whether HD-AOS occurs in this composition range or not. The laser pulse width used to test for AOS varies from 100 fs to 240 fs, while the laser spot diameter varies from 50–80 μm . No qualitative change in the switching behavior was observed for these samples using these laser parameters. It can also be seen from Fig. 6(a) that HD-AOS is not as

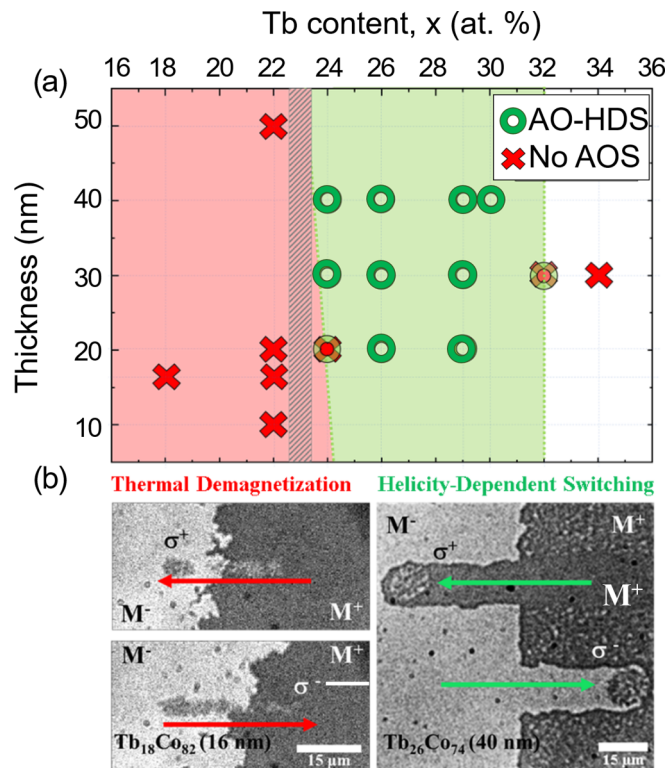


FIG. 6. (a) Diagram showing the switching behavior as a function of sample thickness and composition. The red and green areas correspond to pure thermal demagnetization and multishot helicity-dependent switching, respectively. In the white region, out-of plane domain contrast can no longer be resolved with our microscope. The grey hashed area corresponds to the compensation point x_{comp} . (b) Helicity-independent thermal demagnetization as seen in the red region (left) and HD-AOS as seen in the green-shaded region of the diagram (right).

strongly dependent on sample thickness as it is on the sample composition. Comparing these observations with Fig. 3, which shows that at room temperature $x_{\text{comp}} \sim 23$ at. %, we can conclude that HD-AOS is observed only for samples with $x > x_{\text{comp}}$, i.e., for the samples with $T_{\text{comp}} > 295$ K. This finding agrees with other studies, e.g., by Alebrand *et al.* [11] and Mangin *et al.* [12], that have investigated switching in TbCo alloys as a function of Tb content, and found HD-AOS for samples for which $300 \text{ K} < T_{\text{comp}} < T_{\text{Curie}}$, where T_{Curie} is the Curie temperature. Thus the film composition, with respect to the x_{comp} , clearly has a strong impact on the possibility of achieving HD-AOS.

Some representative results of multipulse switching in $\text{Tb}_{26}\text{Co}_{74}$ films of two different thicknesses are summarized in Fig. 7. A specific threshold fluence, below which no contrast changes, i.e., no switching or demagnetization, can be observed for all investigated samples (dependence of threshold fluence for samples with different Tb content is shown in the Supplemental Material [37], Fig. 9). Therefore, in Fig. 7 we show only the results of measurements performed with a fluence slightly above the threshold fluence. A demagnetized region, clearly resolvable only at a fluence much higher than the threshold fluence, appears with a single laser shot for 40-nm-thick film. As the incident fluence is increased, this

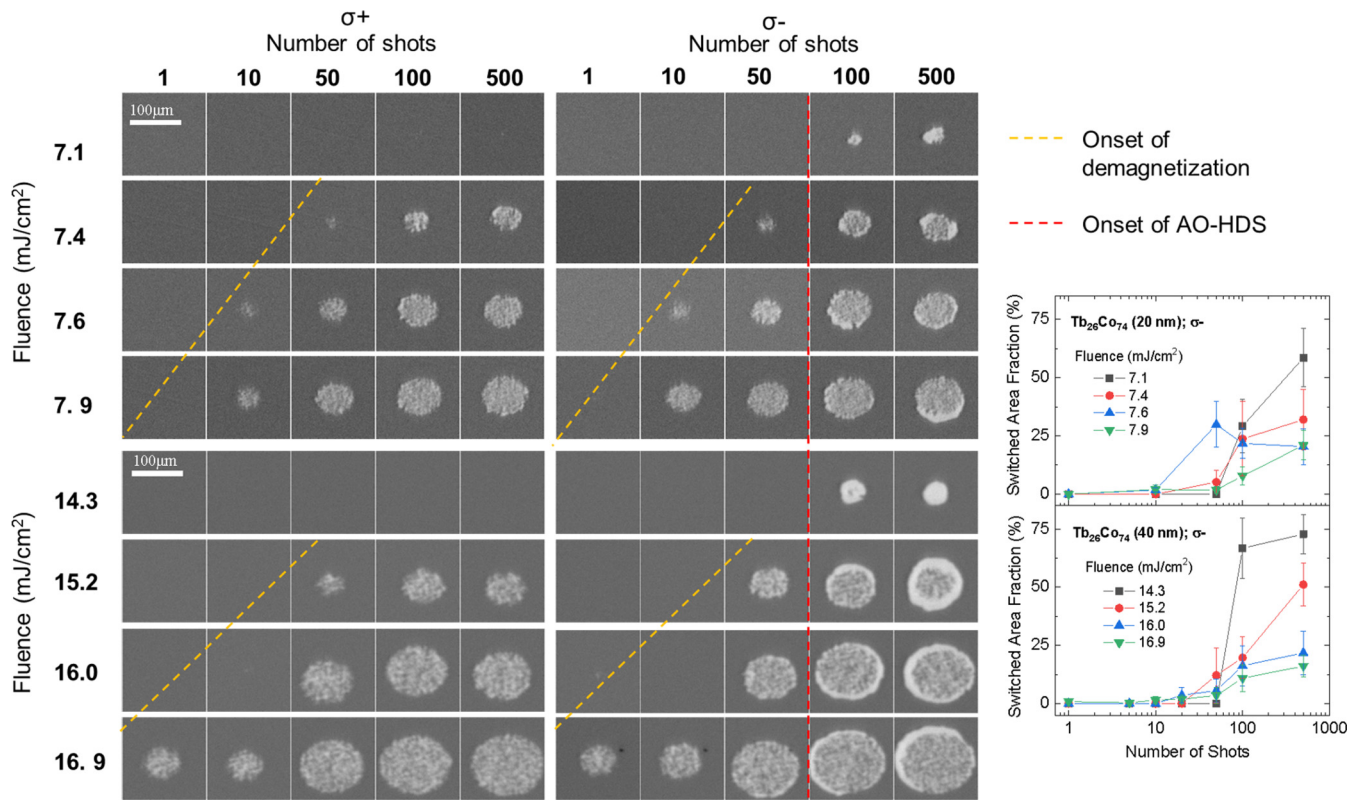


FIG. 7. Observation of HD-AOS dependence on fluence and number of shots in multishot experiments on $Tb_{26}Co_{74}$ films of thickness (a) 20 and (b) 40 nm in response to a train of pulses incident at the same spot on the sample. A ring of switched magnetization appears outside the central demagnetized region for the helicity σ^- for bursts containing 100 or more pulses. The inset shows the efficiency of AOS of magnetization as a ratio of the switched area to the total laser-affected area as a function of number of shots for different fluence for σ^- helicity.

demagnetized region grows larger but no HD-AOS is seen for a single shot of any fluence, relating to recent observations made by Quessab *et al.* [52]. This behavior is also seen for trains of 10 and 50 pulses incident on the sample. Due to the larger number of shots, the demagnetized region grows larger and becomes resolvable for even smaller fluences. The yellow dashed line illustrates how the demagnetization region becomes resolvable at lower fluences for a larger number of shots. The combination of the fluence and number of shots determines the heat deposited to the sample, which in turn determines the size of the demagnetized region. However, no HD-AOS, only demagnetization, is observed below 100 shots, irrespective of the incident fluence, for either of the samples studied here.

Close to the threshold fluence, pure helicity-dependent switching is observed for all the samples, i.e., for one helicity, a complete reversal of magnetization is observed without any central demagnetized region, whereas no contrast change is induced by the opposite helicity. These results further elucidate the effect of film thickness on the switching behavior. Though the film thickness does not play a crucial role in determining the presence of HD-AOS for the samples investigated in this study, it definitely affects the quality of switching. While both 20- and 40-nm-thick $Tb_{26}Co_{74}$ samples show an outer rim of HD-AOS outside the demagnetized region, the rim is much more prominent, thicker and homogeneously shaped for the 40-nm-thick films. This further supports the

observation of better quality of switching for thicker films, possibly due to better heat dissipation upon illumination with a laser pulse, observed in beam sweeping experiments [53].

Results of the beam-sweeping experiments on the 20-nm- and 40-nm-thick $Tb_{26}Co_{74}$ films are shown in Fig. 8, which illustrates the dependence of the observed HD-AOS on the beam sweeping speed. It shows how, on increasing the sweeping speed, the switched area transitions from a homogeneously switched region to discontinuously switched areas. This supports the multishot switching mechanism further. A higher speed correlates to fewer shots per unit length (since the repetition rate of the shots is constant but the beam moves

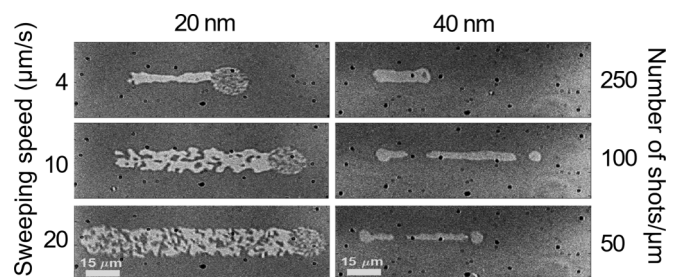


FIG. 8. Sweeping speed dependence of HD-AOS in beam-sweeping experiments on $Tb_{26}Co_{74}$ films of (left) 20- and (right) 40-nm thickness.

over the sample faster) and thus the discontinuity in switching can be explained due to a fewer number of shots and thus smaller fluence received by a unit area. It can be seen from the results obtained for two film thicknesses that the fraction of the laser-swept area that undergoes homogeneous switching is larger for 20-nm thickness as compared to 40 nm, for the same film composition.

We conclude that thickness does not play a crucial role in determining the switching behavior for the 20- to 40-nm-thick films. El Hadri *et al.* [17] studied the role of thickness in HD-AOS in TbCo films for a thickness range of 1.5 nm to 20 nm. In this range, they found that the switching behavior was strongly influenced by the film thickness. This was attributed to the sharp variation of domain sizes as a function of film thickness in this critical regime. Their calculations show that the variation of domain size with film thickness, is not as drastic in the range of 20 – 40 nm. The size of a magnetic domain was considered as one of the criteria for the observation of HD-AOS in both ferromagnetic Co/Pt and Co/Ni films, as well as in ferrimagnetic TbCo alloys of certain thicknesses [17]. More specifically, it was discussed that HD-AOS can be achieved in films of thickness 1.5 – 6 nm with 8 to 15 at. % of Tb, or films of thickness 3.5 – 20 nm with 15 – 30.5 at. % of Tb, and defined a criterion for the observation of AOS to be the equilibrium domain size after laser-induced heating which becomes larger than the laser spot size. Our findings show that HD-AOS can be achieved using suitable laser pulses in thicker TbCo films, which have a composition above the compensation point, and have randomly shaped domains with a broad size distribution from around 40 μm for 20-nm-thick film with 24 at. % Tb to 10 μm and smaller domains for 30-nm-thick films with 24 – 28 at. % Tb. The domain sizes are either comparable or smaller, depending on the composition (and thickness), with our laser spot size of 50 – 80 μm . We observed that even the sample with a composition below the x_{comp} , i.e., 20-nm-thick Tb₁₈Co₈₂, has an average domain size within the range of the domain sizes of the samples with compositions larger than the compensation point. However, it exhibits only helicity-independent thermal demagnetization and not a HD-AOS. Therefore, in our study, the domain size with respect to the laser spot size does not appear to be a determining parameter for the observation of AOS. Instead, we explain the observation of helicity-dependent AOS in our films with the help of theoretical calculations of the IFE.

E. *Ab initio* calculations of the inverse Faraday effect

The *ab initio* computed frequency-dependent IFE constants for the Tb and Co atoms are shown in Fig. 9. Note that in the here-studied excitation geometry the wave vector of the circularly polarized light is parallel to the TbCo magnetization, to simulate a TbCo film with PMA and illuminated at normal incidence. We adopt as a model system the ordered ferrimagnetic Tb₃₃Co₆₇ compound, whose composition is on the edge of the AOS region shown in Fig. 6(a). In the *ab initio* calculations, the Tb atoms have a 9.1 μ_B magnetic moment ($\mu_s + \mu_l$) and the Co atoms an antiparallel 1.3 μ_B moment. Further, we mention that to obtain the laser-induced moment per atom the results given in Fig. 9 are to be multiplied by the unit cell volume and by I/c (for details, see Ref. [28]).

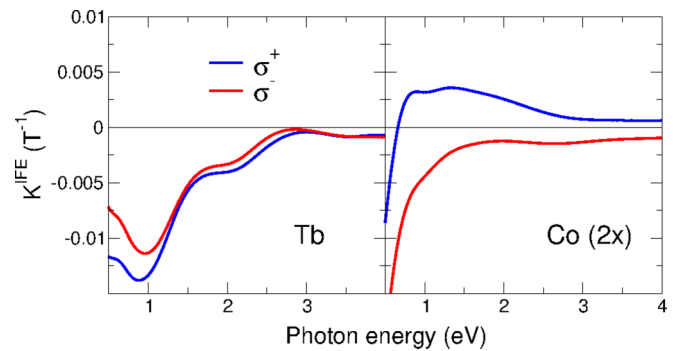


FIG. 9. *Ab initio* computed helicity- and frequency-dependent IFE constants K^{IFE} of Tb and Co atoms in TbCo₂. Note that the values for Co are multiplied by a factor of two.

As can be directly recognized, the induced magnetic moment on the Tb atom displays practically no helicity dependence for a photon energy of 1.55 eV. Consequently, on the Tb atom there will be a laser-induced magnetic moment but it will be antiparallel to the Tb atomic moment irrespective of the light's helicity. This in turn implies that helicity-dependent switching of the large Tb moment is improbable. The situation is, however, different for the Co atom. At around 1.55 eV photon energy, the induced magnetic moment is strongly helicity dependent and, importantly, changes signs when the helicity is reverted, which is favorable for HD-AOS. The helicity-dependent moments induced on the Co atoms can initiate a magnetization reversal but, as its influence is small (about ± 0.05 to $\pm 0.10 \mu_B$ for the here-used experimental fluences), it requires the accumulation of many laser shots in a stochastic process to bring about the magnetization switching. To study the full process of both laser-induced demagnetization and switching, it would be required to perform magnetization dynamics simulations (see, e.g., Refs. [9,29]).

Our first-principles calculations hence show that in particular the helicity-dependent laser-induced moments on the Co atoms are responsible for causing HD-AOS in TbCo, refining recently reported switching mechanisms [52]. A complete picture of helicity-independent AOS and HD-AOS in RE-TM alloys, however, still needs to be achieved. Thus far, helicity-independent, single-shot switching was unambiguously observed for GdFeCo (with $\sim 8\%$ Co) [1]. Recently, single pulse switching was also observed for a Gd/Co bilayer [15]. An ingredient for this switching behavior could be the relatively slow response of the Gd 4*f* magnetic moment upon laser excitation, since the deep-lying 4*f* electrons are not excited by the pump laser, in contrast to the instantaneous excitation of spin-polarized itinerant 3*d* electrons. Recent magnetic dichroism in photoemission experiments measured a slow demagnetization behavior of the Gd 4*f* moment in pure Gd (~ 14 ps) but a subpicosecond demagnetization of the 4*f* moment in Tb [54,55]. This gives an indication that the different intrinsic response times of the 4*f* moments in Gd and Tb could play a role for the AOS. However, XMCD experiments performed on Gd and Tb observed both a fast and a slow 4*f* demagnetization in Gd and in Tb metal [56]. López-Flores *et al.* [57] used XMCD to measure the element-dependent demagnetization rates in GdCo and TbCo alloys

and found subpicosecond demagnetization rates for both Gd and Tb. These different probing techniques thus provide rather disparate values for the $4f$ demagnetization rates and the origin of this discrepancy will need to be clarified to resolve fully why these RE-TM alloys exhibit a distinct AOS behavior. To complicate the matter further, very recently a first observation of single shot AOS was reported for Tb/Co multilayer stacks [7]. Such behavior is, however, not found in our investigation of amorphous TbCo alloys and was neither found in previous studies [3,20].

IV. DISCUSSION AND CONCLUSIONS

We determined the composition and thickness ranges where the $\text{Tb}_x\text{Co}_{100-x}$ films exhibit PMA, reporting that the magnetization compensation point at room temperature is around 23 at. % Tb for the samples deposited onto a fused silica or onto Al_2O_3 buffer layers, while the samples deposited onto $\text{Al}_{80}\text{Zr}_{20}$ have a compensation point around 20 at. % Tb. When deposited onto amorphous layers such as fused silica and alumina, $\text{Tb}_x\text{Co}_{100-x}$ films are amorphous, whereas there are signatures of nanocrystallite formation in the films deposited onto polycrystalline Au buffer layers.

We further explored our parameter space for an optimal region for observation of HD-AOS. We find that $\text{Tb}_x\text{Co}_{100-x}$ alloys which have $x > x_{\text{comp}}$ at room temperature, deposited directly onto fused silica substrates exhibit HD-AOS. For these amorphous films, the sample composition is the most important parameter determining the switching behavior, and HD-AOS was observed for the films with compositions in the range of 24 – 30 at. % of Tb content, while the remaining samples with lower Tb content show thermal demagnetization. The samples with $x > 32$ at. %, exhibit IPMA and therefore no AOS was observed due to our experimental setup, configured for the out-of-plane magnetization switching measurements. Helicity dependence shows up in the sample's response to a train of ultrashort laser pulses only after 50 – 100 shots, with a weak dependence on the applied laser fluence. The film thickness furthermore has a limited influence on HD-AOS, only in terms of raising the threshold fluence, above which either HD-AOS or demagnetization is observed, and by improving the quality of the observed HD-AOS. Lastly, our first-principles calculations explain how the AOS can occur in TbCo. We showed that circularly polarized laser light induces a helicity-dependent magnetic moment on the Co atoms that in particular can trigger the HD-AOS in a cumulative multishot process.

This study shows that ferrimagnetic $\text{Tb}_x\text{Co}_{100-x}$ films in a suitable range of compositions can exhibit HD-AOS under a

range of controlled experimental parameters, such as a laser fluence and sweeping speed. We also show how to control properties like the coercive field, magnetization switching behavior, and room temperature magnetization compensation point, while maintaining an out-of-plane magnetic anisotropy by growing these layers onto a variety of buffer layers. Our observations underline the potential of the combination of TbCo with suitable plasmonic materials to fabricate hybrid magnetoplasmonic architectures [34], possibly enhancing the magneto-optical activity and ultrafast AOS, as was recently shown by Liu *et al.* [50] using patterned Au antennas onto a TbFeCo film or Cheng *et al.* [51] for Co/Pt with Au. Such architectures are potential candidates for the future magnetic memory storage devices based on optomagnetic effects [58].

The data that support this study are available via the Zenodo repository [59].

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