Design of Amorphous Tb$_x$Co$_{100-x}$ alloys for All-Optical Magnetization Switching

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(Dated: February 19, 2020)

Abstract

Amorphous Tb$_x$Co$_{100-x}$ 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 the light-induced magnetization switching. In this work, we investigate the magnetic, optical and magneto-optical properties of various Tb$_x$Co$_{100-x}$ thin film alloy compositions. We report on the effect the choice of different seeding layers has on the structural and magnetic properties of Tb$_x$Co$_{100-x}$ layers. We also demonstrate that for a range of alloys with Tb content of 24-30 at.%, helicity dependent all-optical switching of magnetization can be achieved, albeit in a multi-shot framework. Our study provides an insight into material aspects for future potential hybrid magneto-plasmonic TbCo-based architectures, where enhanced light-induced magnetization switching might be achievable.

I. INTRODUCTION

The development of magnetic memory storage devices was accelerated by advancements in laser technology 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 [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, and even ferromagnetic multilayers [7–11], e.g. Pt-sandwiched Co films [12], to the exploration of the parameter space for the observation of AOS. The latter includes material compositions [13, 14] and laser pulse parameters, such as fluence [15], helicity [16, 17], and pulse duration [18]. A range of both experimental and theoretical works were performed in order to explain the mechanism of AOS: ultrafast heating [19], numerical studies investigating different spin coupling strengths [20], mean field model of relaxation dynamics for ferrimagnets [21]. The switching mechanism is relatively well understood in GdFeCo alloys, where ‘single-shot’ helicity-independent magnetization reversal has been explained as a consequence of the onset of a transient ferromagnetic state. This happens due to different demagnetization rates for the Gd and Fe(Co) sublattices followed by an exchange interaction mediated reversal [22–25]. However, apart from GdFeCo or Co/Pt layers, such a ‘single-shot’ behavior has not been observed in other materials, even in other ferrimagnetic alloys, which instead show all-optical helicity-dependent switching (AO-HDS) in response to a train of multiple circularly-polarized laser pulses. In particular, a study which investigated and contrasted the mechanism of AOS in TbCo and Co/Pt with that in GdFeCo using static magneto-optical imaging and transport measurements based on the Hall effect, reported that switching in TbCo and Co/Pt proceeded via two distinct steps: helicity-independent demagnetization followed by helicity-dependent magnetization recovery [17]. 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 anisotropy in this material, which may pose problems for long-term information retention as well as the down-scaling of the memory bit size. In this respect, the high spin-orbit coupling of Tb renders TbCo a promising material for the AOS, if optimized for its efficiency. Both TbFe and TbCo alloys exhibit larger anisotropy than GdFe and therefore could be more relevant for spintronics-based applications [26].

The magnetic properties of amorphous ferrimagnetic RE-TM Tb$_x$Co$_{100-x}$ alloys are highly tunable. It has been theoretically predicted that Tb$_x$Co$_{100-x}$ could exhibit (due to laser heating) thermally-induced magnetization switching over a wide range of atomic compositions, thus attracting attention as a suitable material for the preparation of the building blocks of non-volatile memory elements [26]. Also, TbCo alloys can exhibit perpendicular magnetic anisotropy [27–29], which makes this alloy attractive since an out-of-plane external magnetic field is not required, allowing for the denser packing of magnetic

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memory elements. The Tb and Co sub-lattices interact anti-ferromagnetically, but their magnetic moments are uncompensated, giving rise to a non-zero net magnetic moment. Starting from a TM-magnetic moment dominated condition at a given temperature, reduction of the Co content in the Tb$_x$Co$_{100-x}$ alloy increases the coercive field and decreases the saturation moment until no net magnetic moment exists. This point is called the magnetization compensation point $x_{\text{comp}}$, 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 $T_{\text{comp}}$ [29].

In this work we present results of a study of magnetic properties and AO-HDS behavior of amorphous RE-TM Tb$_x$Co$_{100-x}$ alloys. We investigate the effect of the atomic ratio of Tb:Co, the thickness of the films and the underlying buffer layers, as well as the influence of the laser pulse characteristics (fluence, number of shots and sweeping speed) on the AOS performance.

II. MATERIALS AND METHODS

Amorphous Tb$_x$Co$_{100-x}$ alloys were prepared at room temperature in an ultrahigh vacuum DC magnetron co-sputtering system, from elemental Tb and Co targets and using Ar as a sputtering gas. The sample holder was rotating during deposition in order to ensure film thickness and composition uniformity. The elemental composition of the samples was investigated by ion beam analysis at the Tandem Laboratory at Uppsala University, using the 5-MV NEC-5SDH-2 tandem accelerator [30]. Rutherford backscattering spectrometry (RBS) and particle induced X-ray emission (PIXE) were employed using 2.0 MeV He$^+$ as a primary beam. More details about the sample preparation and elemental characterization can be found in the Supplementary Information and in Refs. [31, 32]. Films were additionally characterized using X-ray scattering techniques. X-ray reflectivity (XRR) measurements were performed in order to determine layer thickness within the thin film structures. In the data presented in figures we refer to the nominal, i.e. as-calibrated, thickness of the films. The actual thickness was 2 - 4 nm smaller than the nominal one (see XRR data in the Supplementary information). The atomic structure of the films was obtained using grazing incidence X-ray diffraction (GIXRD) measurements in order to determine whether layers are amorphous or crystalline.

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 Tb$_x$Co$_{100-x}$. We measured 20 - 40 nm thick Tb$_x$Co$_{100-x}$/Al$_2$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 for 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 [33, 34], 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 Supplementary Information.

Static magnetic and magneto-optical properties of the Tb$_x$Co$_{100-x}$ films were characterized 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 remnant magnetization state of Tb$_x$Co$_{100-x}$ films was imaged using Kerr microscopy. We used a Kerr microscope with a white light source and applied the magnetic field perpendicular to the sample plane. In order 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 analysed using a pair correlation function (PCF). 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 [35], 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.

In order to investigate AO-HDS of magnetization, static magneto-optical imaging was performed. 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× 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 AO-HDS, 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 multi-shot measurements, the pulse width at the sample was 90 fs and incident at 15° 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° to the sample normal. A translation stage
was used to control the sweep speed for the sweeping beam measurements.

III. RESULTS

A. Ion beam analysis characterization

A typical RBS spectrum from Tb\textsubscript{18}Co\textsubscript{82} sample studied in this work 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 better than 1%. The elemental composition was determined for samples deposited under different deposition conditions in order 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 ($\approx 0.2$ 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.

B. Structure of Tb\textsubscript{x}Co\textsubscript{100-x} films

The structure of the fabricated Tb\textsubscript{x}Co\textsubscript{100-x} films prepared on fused silica substrates, with and without a Au buffer layer, was investigated employing GIXRD measurements. The Au buffer layers were chosen in this study in order to investigate influence on the structure and thus possibility of directly combining Tb\textsubscript{x}Co\textsubscript{100-x} alloys with typical plasmonic materials such as Au, a prerequisite for the fabrication of future Tb\textsubscript{x}Co\textsubscript{100-x}-based magneto-plasmonic architectures.

A typical GIXRD diffractogram for two Tb\textsubscript{30}Co\textsubscript{70} films, one prepared on a fused silica substrate and the other- on a Au buffer layer, are shown in Fig. 2. The film prepared directly onto a fused silica substrate exhibits a single, low intensity broad peak at around $2\theta = 22^\circ$, that can be associated with amorphous SiO\textsubscript{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 $2\theta = 22^\circ$, similarly 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 Tb\textsubscript{30}Co\textsubscript{70} film.

It was shown in the work by Frisk et al. [28] that amorphism of Tb\textsubscript{x}Co\textsubscript{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\textsubscript{2}O\textsubscript{3}, grow amorphous (since narrow high intensity peaks in the GIXRD were not observed), 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. [38]. Crystallite sizes, computed using Scherrer’s equation, are 97.9 and 58.7 Å for Au and Tb\textsubscript{30}Co\textsubscript{70} layers, respectively. To further confirm amorphous structure of the Tb\textsubscript{x}Co\textsubscript{100-x} films, we performed extended X-ray absorption fine structure (EXAFS) measurements of a Tb\textsubscript{2}Co\textsubscript{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 the Supplementary information.

C. Optical characterization

The wavelength dependence of the refractive index, $n$, and extinction coefficient, $k$, extracted from the ellipsometry measurements, are shown in Fig. 3. The measured optical constants are $n = 2.32$ and $k = 0.01$ at 632.8 nm for the Tb\textsubscript{x}Co\textsubscript{100-x} films on fused silica substrates.
metric data, is summarized in Fig. 3 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 [39]. It appears that there is no significant dependence on the Tb content for samples with a Tb content above the $x_{\text{comp}}$. 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 $x > x_{\text{comp}}$. Once again, the optical constants determined here have been targeted having in mind the potential of Tb$_x$Co$_{100-x}$ films in magneto-plasmonic architectures (see i.e., work by Liu et al. [40]). For the proper electromagnetic design and simulations of these, knowledge of these optical constants is of paramount importance.

D. Magnetic and chemical characterization

We investigated how the magnetic properties of Tb$_x$Co$_{100-x}$ films are affected by the different buffer layers on which they are deposited and imaged the remanent magnetization state. The effect of various buffer layers on the coercive field of a 18 nm thick Tb$_{18}$Co$_{82}$ film is illustrated in Fig. 4. The coercive field is largest for samples prepared on an Al$_{80}$Zr$_{20}$ buffer layer while the films deposited on a 20 nm thick Au buffer layer appear to have a reduced coercive field. This can be related to the structure of the Tb$_x$Co$_{100-x}$ films: Al$_{80}$Zr$_{20}$ is well known to promote the amorphous growth of the films, while a polycrystalline Au buffer promotes nanocrystalline formation in TbCo films at the interface (see Fig. 2) [38]. However, an increase in the coercive field can be observed when a Tb$_{18}$Co$_{82}$ film is deposited onto a hybrid buffer of Au/Al$_2$O$_3$ so that Tb$_{18}$Co$_{82}$ has an interface with amorphous Al$_2$O$_3$. This demonstrates how the higher degree of amorphicity allows for increased coercivity in Tb$_x$Co$_{100-x}$ alloys.

FIG. 2. Diffractograms of a Tb$_{30}$Co$_{70}$ sample deposited onto a fused silica substrate (red points) and onto a Au buffer layer (grey points), both capped with Al$_2$O$_3$. Diffractograms were deconvoluted into Lorentzians, given by the solid black lines, and the intensity was normalized to the SiO$_2$ peak. The black measurement points have been shifted upwards for clarity.

FIG. 3. 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 GenX fits.

FIG. 4. Coercive field dependence for 20 nm thick Tb$_{18}$Co$_{82}$ films deposited on different buffer layers.
We further analyzed how the coercive field $\mu_0 H_c$ changes by varying composition and sample thickness. We varied the Tb content in the Tb$_x$Co$_{100-x}$ films of 20, 30 and 40 nm in thickness, deposited on untreated fused silica substrates and on a hybrid Au(20nm)/Al$_2$O$_3$ buffer layers. The results are summarized in Fig. 5. For films grown on the untreated silica substrates, the coercive field 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 $x_{\text{comp}}$ at room temperature. Similar values of Tb$_x$Co$_{100-x}$ $x_{\text{comp}}$ at room temperature were reported in work by Alebrand et al. [9]. 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 perpendicular, magnetic anisotropy (PMA). Our investigations also show that, reduction of the thickness of TbCo films to below 10 nm, results in the emergence of in-plane magnetic anisotropy at the expense of the desired PMA (see Supplementary information). Similarly to observations in Fig. 4, the Tb$_x$Co$_{100-x}$ films prepared on hybrid Au(20nm)/Al$_2$O$_3$ buffer layers exhibit larger coercive fields than their counterparts prepared directly on fused silica substrates.

In relation to the seed layer effect investigation, we also measured samples with 15, 18 and 22 at.% Tb prepared onto Al$_{80}$Zr$_{20}$ 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 Supplementary Information). This indicates that the compensation point for films prepared onto Al$_{80}$Zr$_{20}$ 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. [28]. These results demonstrate the importance of an underlying buffer layer for the observed magnetic properties of the Tb$_x$Co$_{100-x}$ films.

We imaged the magnetic domain structure of the 20 nm thick Tb$_x$Co$_{100-x}$ films, deposited on fused silica substrates, using Kerr microscopy and XPEEM. Kerr microscopy images show a size distribution for the magnetic domains (see the Supplementary information). Magnetic contrast maps were further recorded at the Co $L_{2,3}$ and the Tb $M_5$ edges are shown in Fig. 6(c) and (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 alloy. X-ray absorption (XA) spectra shown in Fig. 6(a) and (b), reveal a pure and metallic sample without traces of oxidation.

E. Static Magneto-optical Imaging: All-optical Helicity Dependent Switching

TbCo samples with varying Tb content, grown directly on fused silica substrates, were investigated for AOS behavior. Single-shot helicity-independent switching of

![FIG. 5. Coercive field dependence on Tb$_x$Co$_{100-x}$ composition for samples deposited on fused silica substrates (illustrated as circles), on mixed silica/Au(20nm)/Al$_2$O$_3$ (3 nm) buffer structures (triangles) and on Al$_{80}$Zr$_{20}$ (3nm) buffer layers (squares). Black, red and empty symbols correspond to $t = 20$, 30 and 40 nm thickness of Tb$_x$Co$_{100-x}$ layer, 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 $x_{\text{comp}}$ for the sample series prepared directly on fused silica substrate and onto Al$_{80}$Zr$_{20}$ (3nm) buffer layers, respectively. Measurements were performed in PMOKE configuration, with incident light of 530 nm wavelength.](image)

![FIG. 6. 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 Tb$_{24}$Co$_{76}$ film obtained by combining XPEEM with XMCD at the (c) Co $L_3$ and the (d) Tb $M_5$ edges.](image)
magnetization was not observed in any of the samples studied. However, a range of samples do exhibit a multi-shot AO-HDS under suitable laser pulse parameters, such as fluence and sweeping speed (for details see the Supplementary information). The range of concentration and thickness, where AO-HDS was observed for these samples, is plotted in the diagram of Fig. 7(a). Below 24 at.\% Tb content, thermal demagnetization is always observed. This is shown in the left panel of Fig. 7(b), where both of the incident helicities, $\sigma^+\text{ and } \sigma^-$, 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 AO-HDS, as seen in the right panel of Fig. 7(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 $x>32\text{ at.\%}$, domain contrast cannot be observed with the polar Faraday imaging geometry, due to the shifting of easy out-of-plane magnetization axis towards the in-plane direction. Hence, it is unresolved whether AO-HDS 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 40 to 160 $\mu$m. No qualitative change in the switching behavior was observed for these samples using these laser parameters. It can also be seen from Fig. 7(a) that AO-HDS is not as strongly dependent on sample thickness as it is on the sample composition. Comparing these observations with Fig. 5, which shows that at room temperature $x_{\text{comp}} \sim 23\text{ at.\%}$, we can conclude that AO-HDS 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. [9], Mangin et al. [10], that have investigated switching in TbCo alloys as a function of Tb content, and found AO-HDS for samples for which $300 \ K < T_{\text{comp}} < T_{\text{Curie}}$, where $T_{\text{Curie}}$ is the Curie temperature. Thus the film composition, with respect to the $x_{\text{comp}}$, clearly has a strong impact on the possibility of achieving AO-HDS.

Some representative results of multi-pulse switching in Tb$_{26}$Co$_{74}$ films of two different thicknesses are summarized in Fig. 8. A specific threshold fluence, below which no contrast changes, i.e. no switching or demagnetization, can be observed for all investigated samples. Therefore, in Fig. 8 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 demagnetized region grows larger but no AO-HDS is seen for a single shot of any fluence. 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 AO-HDS, 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 AO-HDS 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 AO-HDS 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 (see Supplementary information) [41].

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. [14] studied the role of thickness in AO-HDS 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. In the work by El Hadri et al. [14] the size of a magnetic domain was considered as one of the criteria for the observation of AO-HDS in both ferromagnetic Co/Pt and Co/Ni films, as well as in ferrimagnetic TbCo alloys of certain thicknesses. More specifically, they showed that AO-HDS 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. Our findings show that AO-HDS can be achieved in thicker TbCo films, which have randomly shaped domains of sizes from around 15 µm for 20 nm to submicron size domains for 40 nm thick films.

IV. DISCUSSION AND CONCLUSIONS

We determined the composition and thickness ranges where the Tb$_x$Co$_{100-x}$ films exhibit perpendicular magnetic anisotropy, 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$_2$O$_3$ buffer layers, while the samples deposited onto Al$_{80}$Zr$_{20}$ have a compensation point around 20 at.% Tb. When deposited onto amorphous layers such as fused silica and alumina, Tb$_x$Co$_{100-x}$ films are amorphous, while 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 AO-HDS. We find that
Tb$_2$Co$_{100-x}$ alloys which have $x > x_{\text{comp}}$ at room temperature, deposited directly onto fused silica substrates exhibit AO-HDS. For these amorphous films, the sample composition is the most important parameter determining the switching behavior, and AO-HDS 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 in-plane magnetic anisotropy and therefore no AOS was observed due to our experimental setup, configured for the out-of-plane magnetization switching measurements. The mechanism for AO-HDS in Tb$_2$Co$_{100-x}$ samples investigated here is not a single-shot helicity-independent mechanism like in GdFeCo, but rather a cumulative multi-shot mechanism similar to the Co/Pt multilayers. Helicity dependence shows up in the sample’s response to a train of ultrashort laser pulses only after 50 - 100 shots, irrespective of the applied laser fluence. The film thickness has a limited influence on AO-HDS, only in terms of raising the threshold fluence, above which AO-HDS/demagnetization is observed, and by improving the ‘quality’ of the observed AO-HDS.

This study shows that ferrimagnetic Tb$_2$Co$_{100-x}$ films in a suitable range of compositions can exhibit AO-HDS under a range of controlled experimental parameters, such as a laser fluence and sweeping speed. We also show how to control the properties like the coercive field, switching behavior and room temperature compensation point, while maintaining an out-of-plane magnetic anisotropy by growing these layers onto a variety of buffer layers. These observations show the potential of the combination of TbCo with suitable plasmonic materials in order to fabricate hybrid magneto-plasmonic architectures, possibly enhancing the magneto-optical activity and ultrafast AOS, as was recently shown by Liu et al. [40] using patterned Au antennas onto a TbFeCo film. Such architectures are potential candidates for the future magnetic memory storage devices based on opto-magnetic effects.

The data that support this study are available via the Zenodo repository [42].

ACKNOWLEDGMENTS

The authors acknowledge support from the Knut and Alice Wallenberg Foundation project "Harnessing light and spins through plasmons at the nanoscale" (2015.0060) and the Swedish Foundation for International Cooperation in Research and Higher Education. This work is part of a project which has received funding from the European Union’s Horizon 2020 research and innovation program under grant agreement no. 737093, "FEMTOTERABYTE".

The operation of the Tandem Accelerator Laboratory has been supported by infrastructural grants from the Swedish Foundation for Strategic Research (SSF-RIF14-0053) and the Swedish Research Council (contracts # 821-2012-5144 and # 2017-00646-9).

KM and AK (Andrei Kirilyuk) would like to acknowledge Guido Bonfiglio, Dr. Kihiro Yamada for assistance with experimental setups for imaging and temperature-dependent hysteresis measurements, and Dr. Sergey Semin and Chris Berkhout for technical support. MVM would like to acknowledge Dr. Daniel Primetzhofer for discussions in the ion beam analysis section. AC, GA and VK would like to thank Dr. Maarten Nachttegaal and SuperXAS - X10DA beamline at PSI for providing beam-time for EXAFS measurements, and Sebastian George for help in the beamtime and data analysis, as well as for providing initial samples for AOS testing. Dr. Gunnar K. Pålsson is acknowledged for help with computational aspects of the domain size evaluation and GIXRD.

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