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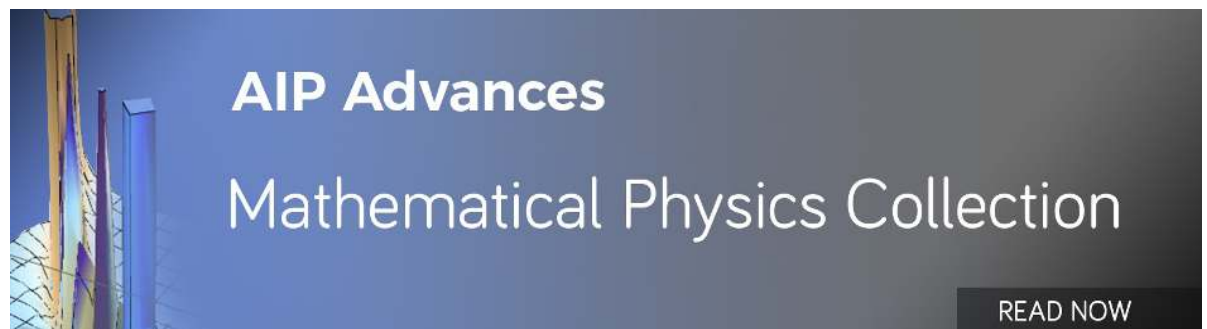
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ABSTRACT

This work reports the development of perpendicular magnetic tunnel junctions, based on a multilayered stack of [Tb/Co] nanolayers, in which the magnetization can be reliably toggled using a single optical pulse. The helicity-independent single-shot switching of the magnetization in the Tb/Co multilayered stack was achieved using either 60 fs-long or 5 ps laser pulses with incident fluences down to 4.7 mJ/cm². The magnetic switching was achieved for a Co-rich composition window of the multilayer corresponding to layer thickness ratios $t_{\text{Co}}/t_{\text{Tb}}$ between 1.3-1.5. This was confirmed for the multilayer alone as well as for the multilayer coupled to aCoFeB electrode, with a structure consisting of CoFeB/Ta/[Tb/Co]_N. The optical switching is preserved even after annealing at 250°C in magnetic tunnel junctions (MTJ) electrodes, exhibiting a tunnel magnetoresistance (TMR) of 41% and RxA value of 150 Ωμm after its integration, measured on unpatterned MTJ stacks. These results represent the first step towards the development of hybrid spintronic photonic systems with fJ switching energies.

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I. INTRODUCTION

Rare earth (RE)- transition metal (TM) alloys and multilayers have been extensively studied due to their potential application in several fields of magnetism, being of particular importance in the field of magneto-optical recording.¹ The magnetic properties of RE-TM thin films, both in amorphous alloy forms and as a multilayered structures have been explored in detail, and previous works have highlighted their strong perpendicular magnetic anisotropy (PMA)² and the possibility to tune the anisotropy constant by varying the relative proportions of the TM and RE constituents.³ In a more recent context, it has been demonstrated that RE-TM-based alloys, such as GdFeCo or TbCo exhibit the phenomena of all-optical switching (AOS),^{4,5} whereby the magnetization can be reversed using femtosecond- or picosecond-long laser pulses,

without the assistance of an external magnetic field. AOS can be classified, depending on the necessary properties of the laser pulses, as helicity-dependent (AO-HDS) or helicity-independent (AO-HIS). These effects have been observed to induce magnetization reversal in different materials either with a single pulse or with multiple consecutive pulses. Different studies have reported that the AOS strongly depends on the magnetic properties of the materials. For example, GdFeCo alloys exhibit both AO-HDS and AO-HIS with single pulses,⁴ whereas, TbCo alloys only displays AO-HDS after exposure to multiple pulses.⁵ Furthermore, as we recently observed deterministic AO-HIS in [Tb/Co]_N multilayers,⁶ this ferrimagnetic system being suitable candidate for integration in a magnetic tunnel junction (MTJ).⁷ We have therefore optimized [Tb/Co]_N multilayer structures magnetically coupled to a CoFeB layer in order to serve as the storage layer in a magnetic

tunnel junction allowing for single-shot AOS through ultrafast laser pulses.

This paper reports the development of a perpendicular MTJ, incorporating a $[\text{Tb}/\text{Co}]_N$ -based multilayered stack coupled to a CoFeB layer in order to form the storage layer of a memory MRAM. A special focus is done on the influence of the annealing temperature on the magnetic properties of the multilayer. It is known that RE-TM systems are sensitive to post-deposition annealing.⁸ However, we demonstrate that the PMA is preserved after annealing at 250°C and the capacity for single-shot all-optical switching is conserved.

II. EXPERIMENTAL DETAILS

Crossed-wedge thickness structures consisting of $\text{Si}/\text{Ta}(30\text{\AA})/[\text{Co}(t_{\text{Co}})/\text{Tb}(t_{\text{Tb}})]_N/\text{Cu}(20\text{\AA})/\text{Pt}(30\text{\AA})$ have been fabricated with different thicknesses of the Tb and Co layers ranging from 6 to 16 Å for Tb and from 7 to 14 Å for Co. The brackets contain the bilayer structure that is repeated N times within the stack. Samples have been grown on thermally-oxidized single-crystal Si(100) wafers by DC magnetron sputtering, using Ar pressure of 2×10^{-3} mbar and a

base pressure of 10^{-8} mbar. Quasi-static magnetic hysteresis loops were measured with an Extraordinary Hall Effect (EHE) set-up, and coercive field (H_c) mappings were obtained with a Kerr magnetometer in polar configuration. The maximum value of the magnetic field in our Kerr set-up is 2.4 kOe. X-ray reflectivity (XRR) measurements were performed to study the influence of annealing on the structural properties of multilayers by measuring the periodicity and the intensity of the Kiessig fringes. To achieve AOS of the magnetization, we used in-situ magneto-optical imaging microscopy in the polar Kerr configuration. The optical pulses at 800 nm were generated by a pulsed-amplified-laser system capable of generating laser pulses of tunable duration on a single-shot basis.

III. RESULTS

Hysteresis loops obtained from different regions of the crossed-wedge samples allowed us to identify the range of thicknesses for which the easy axis of the uniaxial anisotropy is perpendicular to the plane of the layers. Fig. 1a shows the coercive field mapping of the sample as-deposited for different values of the thicknesses t_{Co} and t_{Tb} . In the mapping, red and blue areas represent high and low

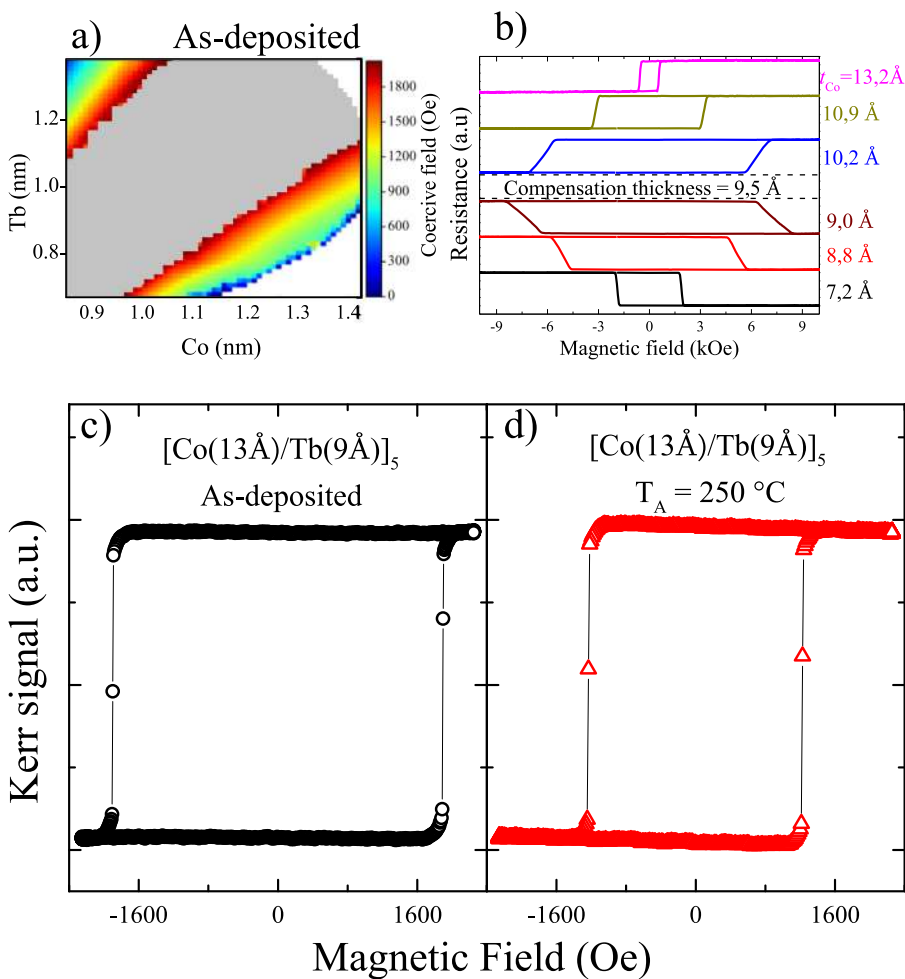


FIG. 1. (a) H_c mapping of $[\text{Co}(t_{\text{Co}})/\text{Tb}(t_{\text{Tb}})]_5$ multilayers as-deposited. The coercivity values were obtained from the $M(H)$ loops measured with H applied perpendicular to the plane of the films. (b) $R(H)$ of $[\text{Co}(t_{\text{Co}})/\text{Tb}(10\text{\AA})]_5$ as a function of t_{Co} across the compensation thickness. (c) Kerr hysteresis loops of $[\text{Co}(13\text{\AA})/\text{Tb}(9\text{\AA})]_5$ obtained from the sample as-deposited and (d) from the sample annealed at 250°C.

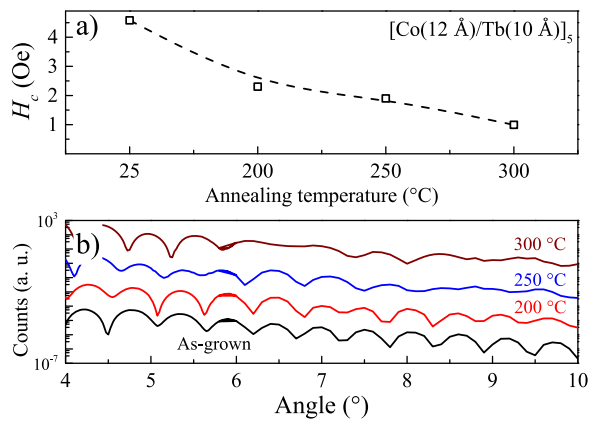


FIG. 2. (a) Coercive field of the multilayer $[\text{Co}(12\text{\AA})/\text{Tb}(10\text{\AA})]_5$ as a function of the annealing temperature obtained from EHE experiments. Dotted line is a guide to the eyes. b) XRR measurements of $[\text{Tb}(10\text{\AA})/\text{Co}(12\text{\AA})]_5$ samples as-deposited and annealed at 200, 250 and 300°C.

values of the coercive field respectively. The grey region in the center indicates the areas of the sample where the maximum external magnetic field available in our setup is not strong enough to reverse the magnetization of the Tb/Co multilayer.

The increase of the coercive field as the composition approaches the 1:1 $t_{\text{Co}}/t_{\text{Tb}}$ thickness ratio indicates that the

magnetic moment compensation of the Co and Tb sublattices at room temperature occurs in the central part of the composition region. This was validated by analysing of the $M(H)$ loops at each side of the compensation region for the samples as grown. EHE hysteresis loops for $[\text{Co}(t_{\text{Co}})/\text{Tb}(10\text{\AA})]_5$ as a function of t_{Co} across the compensation thickness are shown in Fig. 1b in which the inversion of the loops with respect to the magnetic field is due to the fact that the main contribution to the EHE signal is from the Co sublattice. As the ratio of Tb is increased, the Tb sublattice total moment becomes dominant and higher than that of Co sublattice, so that the Co moment then becomes opposite to the saturation field because of the antiferromagnetic coupling between the Tb and Co sublattices. Figures 1c and 1d present the hysteresis loop of $[\text{Co}(13\text{\AA})/\text{Tb}(9\text{\AA})]_5$, for samples as-deposited and after annealing at 250°C respectively. Although the coercive field decreases around 25% for $[\text{Co}(13\text{\AA})/\text{Tb}(9\text{\AA})]_5$ after annealing, the high remanence and the squareness of the hysteresis loop is maintained for most of the Co/Tb thickness ratio's. These results agree with previous reports on the thermal stability of [Co/Tb] multilayers, in which the magnetic anisotropy is strongly affected by annealing, even at relatively low temperatures⁹ ($\approx 200^\circ\text{C}$). Although the PMA is only preserved for annealing temperatures up to 275°C in $[\text{Co}(13\text{\AA})/\text{Tb}(9\text{\AA})]_5$ or $t_{\text{Co}}/t_{\text{Tb}} = 1.44$, the thermal stability strongly depends on the $t_{\text{Co}}/t_{\text{Tb}}$ values. This is clearly observed in samples with $t_{\text{Co}}/t_{\text{Tb}}$ ratios close to the magnetic compensation. For instance, the multilayer $[\text{Co}(12\text{\AA})/\text{Tb}(10\text{\AA})]_5$ ($t_{\text{Co}}/t_{\text{Tb}} = 1.2$) kept a strong PMA and a coercive field of 990 Oe even after annealing at 300°C, as can be seen in Fig. 2a. Several studies attribute this anisotropy loss

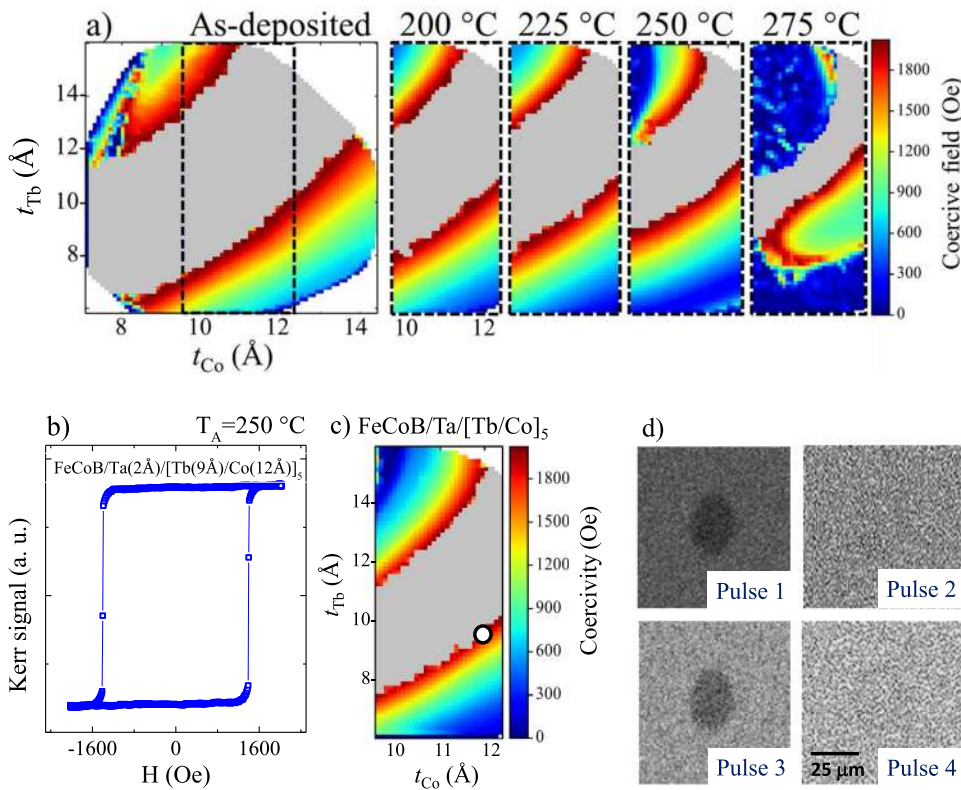


FIG. 3. a) Evolution of the coercivity mappings of the $\text{CoFeB}(13\text{\AA})/[\text{Tb}(t_{\text{Tb}})/\text{Co}(t_{\text{Co}})]_5$ electrode after annealing at different temperatures from 200°C to 275°C. b) $M(H)$ and c) coercivity mapping of the $\text{CoFeB}/\text{Ta}(2\text{\AA})/[\text{Tb}/\text{Co}]_5$ stack. d) Single shot AOS of the electrode after consecutive 5ps-long laser pulses with an incident fluence of $4.7\text{ mJ}/\text{cm}^2$.

to different structural transformations at the interfaces during the annealing¹⁰ such as possible increase of interfacial roughness due to interdiffusion or even structural relaxation modifying the strain within the sample. This was verified with low angle XRR measurements performed in the multilayer with $t_{\text{Co}}/t_{\text{Tb}}=1.2$ annealed at different temperatures as shown in Fig. 2b. As can be observed in the XRR pattern, the amplitude of the oscillating peaks decreases with increasing the annealing temperature and completely vanishes after annealing at 300 °C. This indicates that the interfacial roughness and the long range order in the multilayer is affected by the post-deposition annealing process. This degradation can be correlated with the decrease of the coercive field and can explain the loss of PMA.

To introduce our [Tb/Co]_N system as part of the storage layer in a MTJ we characterized the magnetic coupling between the CoFeB electrode and the [Tb/Co]_N multilayer by growing the multilayer either in direct contact with the CoFeB electrode or through a thin Ta insertion layer. We fabricated crossed-wedge samples with the following composition: MgO/CoFeB(13Å)/[Tb(*t*_{Tb})/Co(*t*_{Co})]₅ and MgO/CoFeB(13Å)/Ta(2Å)/[Tb(*t*_{Tb})/Co(*t*_{Co})]₅. The Co and Tb sublattices are antiferromagnetically coupled in the multilayer. The additional CoFeB electrode can be coupled to the multilayer, either directly to the Co sublattice, or through a bottom Tb layer, to maintain an antiferromagnetic coupling between the CoFeB and the [Tb/Co] multilayer. Results are shown for coupling with Tb as bottom layer in our stack. Coercivity mapping of the MTJ electrode without the ultra-thin layer of Ta are plotted in Fig. 3a. PMA is maintained even when the CoFeB electrode is coupled to the multilayer. Furthermore, the magnetic compensation point at room temperature occurs for the same composition $t_{\text{Co}}/t_{\text{Tb}} \sim 1.1$. The evolution of the coercivity mapping after annealing conditions from 200 °C up to 275 °C, is shown in Fig. 3a, representing the maximum temperature before permanent loss of perpendicular anisotropy is observed. This loss is observed for higher Co composition values after annealing at 275 °C. However PMA is still present at Co compositions closer to compensation. The squareness and the presence of a single step hysteresis loop of the CoFeB(13Å)/Ta(2Å)/[Tb(9Å)/Co(12Å)]₅ stack, as can be seen in Fig. 3b, indicates that the coupling between the CoFeB and the [Tb/Co]₅ is sufficiently strong to insure simultaneous switching of the CoFeB layer and [Tb/Co]₅ ferrimagnet.

Ultrafast AOS of the magnetization of the storage electrode stack was explored in several regions of the CoFeB(13Å)/Ta(2Å)/[Tb/Co]₅ sample after annealing at 250 °C, using both 5 picosecond and 60 femtosecond laser pulses. Magnetization switching was achieved using either ps- or fs-pulses with fluences down to 4.7 mJ/cm² and observed only for Co-rich compositions of the storage electrode. A solid line circle in Fig. 3c indicates the region of the storage electrode illuminated with single laser pulses and Fig. 3d shows Kerr microscopy images obtained after a sequence of 4 single shots on the CoFeB(13Å)/[Tb(9Å)/Co(12Å)]₅ multilayer using 5 ps-long laser pulses. The magneto-optical response to a single laser pulse in different regions of the storage electrode allowed us to identify that the AOS process is possible in a Co-rich composition window corresponding to layer thickness ratios $t_{\text{Co}}/t_{\text{Tb}}$ between 1.3 - 1.5. It is important to mention that [Tb/Co]₅ multilayers without CoFeB electrode also showed ultrafast switching for the same thickness ratios. The ultrafast magneto-optical response was also tested in samples with an increased number of repetitions ($N=15$), but the

incident fluence required to reverse the magnetization increased to 19 mJ/cm², which is about 5 times higher compared to the samples with 5 repetitions.

For the integration into a full MTJ, the optically switchable storage electrode was grown as a top electrode on top of a MgO barrier, using a low coercivity CoFeB layer as a counter electrode. The final stack structure CoFeB(11Å)/MgO/CoFeB(13Å)/Ta(2Å)/[Tb/Co]_N was used to evaluate TMR for MgO tunnel barriers having RxA values from 10 up to 150 Ωμm². The effect of the number of [Tb/Co]_N bilayer repetitions was investigated in full magnetic tunnel junctions. Fig. 4 shows the hysteresis loop of the CoFeB(11Å)/MgO/CoFeB/Ta/[Tb(9.5Å)/Co(12.5Å)]_N multilayers with $N = 5, 10$ and 15 annealed at 250 °C. The hysteresis loop of the samples with 5 repetitions of [Tb/Co] evidences the two magnetization states, corresponding to the magnetization of the CoFeB/Ta/[Tb/Co]_N electrodes and that of the free CoFeB layer. As the number of repetitions increases, the antiferromagnetic coupling of the [Tb/Co]_N multilayer is reduced, as can be seen from

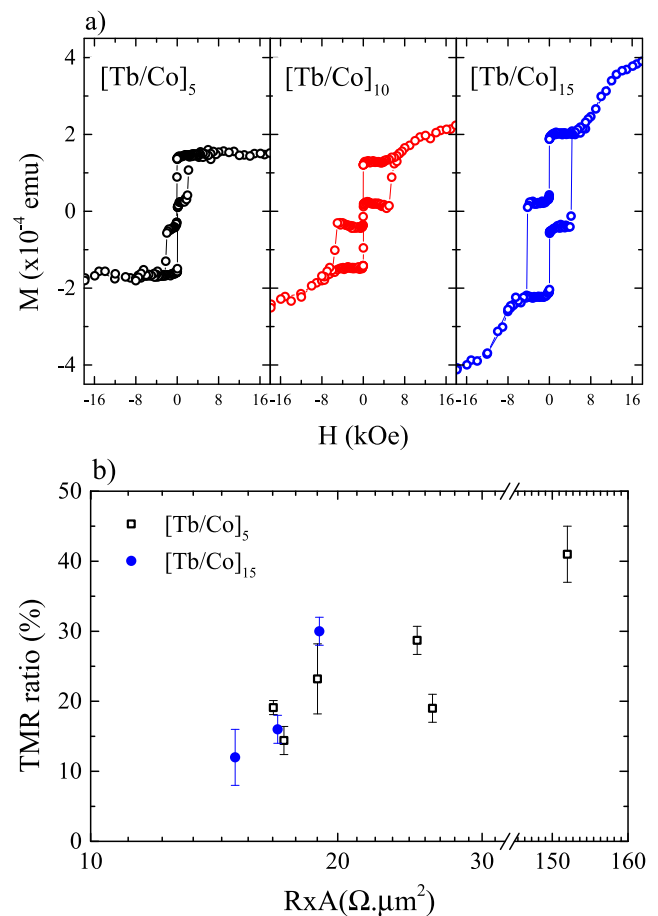


FIG. 4. (a) $M(H)$ of CoFeB/MgO/CoFeB/Ta/[Tb(9.5Å)/Co(12.5Å)]_N for different number of repetitions $N = 5, 10$ and 15 annealed at 250 °C. (b) TMR ratios extracted from CIPT measurements in CoFeB/MgO/CoFeB/Ta/[Tb/Co]_N annealed at 250 °C.

the multilayer saturation observed at higher fields for 10 and 15 repetitions.

Current-in-plane tunneling (CIPT) measurements were performed on fullsheet film samples with 5 and 15 repetitions after annealing at 250°C. The extraction of the TMR values was done by reversing the magnetization of the free layer electrode from parallel to anti-parallel alignment with the external magnetic field. In the samples with 15 repetitions the maximum TMR ratio and R_{xA} value were $\sim 30\%$ and $19 \Omega\mu\text{m}$. On the other hand, samples with 5 repetitions of the [Tb(9.5 Å)/Co(12.5 Å)] bilayer showed higher values of TMR up to 41% for an R_{xA} of $150 \Omega\mu\text{m}$. At lower R_{xA} levels, TMR values ranged from 12% to 30% were obtained for both 5 and 15 repetition samples, as can be seen in Fig. 4b. These values are a clear indication that this multilayer system is a good candidate for integration in spintronic devices allowing for optical switching.

IV. CONCLUSION

We successfully integrated an optically switchable CoFeB-[Tb/Co]_N electrode in a perpendicular anisotropy MTJ. Control of the coercive field value and the perpendicular magnetic anisotropy of the electrode is possible by adjusting the Co/Tb composition in the multilayer using different $t_{\text{Co}}/t_{\text{Tb}}$ thickness ratios. An annealing study showed that antiferromagnetic alignment between the Tb and Co sublattices is maintained even after annealing at 250°C. AOS of the magnetization in the CoFeB/Ta/[Tb/Co]_N storage electrode was observed using both 60fs and 5ps laser pulses with incident fluences as small as 4.7 mJ/cm^2 . This behavior is observed for a Co-rich composition window with Co/Tb multilayer thickness ratios between 1.3 - 1.5. After annealing, TMR signal up to 41% for an R_{xA} value of $150 \Omega\mu\text{m}$ were obtained for a storage

electrode of CoFeB(11Å)/MgO/CoFeB/Ta/[Tb(9.5Å)/Co(12.5Å)]₅. Full stack structures with 15 repetitions of [Tb/Co] showed similar values of TMR. These results pave the way for the development of hybrid spintronic-photonic systems with unique features of THz MTJ switching speeds and fJ switching energies.

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