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Citation for published version (APA):
Hintermayr, J., Li, P., Rosenkamp, R., van Hees, Y. L. W., Igarashi, J., Mangin, S., Lavrijsen, R., Malinowski, G., & Koopmans, B. (2023). Ultrafast single-pulse all-optical switching in synthetic ferrimagnetic Tb/Co/Gd multilayers. *Applied Physics Letters*, *123*(7), Article 072406. https://doi.org/10.1063/5.0161322

DOI:
10.1063/5.0161322

Document status and date:
Published: 14/08/2023

Document Version:
Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Applied Physics Letters

RESEARCH ARTICLE | AUGUST 17 2023

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Special Collection: Ferrimagnetic Spintronics

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Appl. Phys. Lett. 123, 072406 (2023)
https://doi.org/10.1063/5.0161322

Applied Physics Letters ARTICLE pubs.aip.org/aip/apl

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Cite as: Appl. Phys. Lett. **123**, 072406 (2023); doi: 10.1063/5.0161322
Submitted: 9 June 2023 · Accepted: 7 August 2023 ·
Published Online: 17 August 2023

View Online Export Citation CrossMark

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Note: This paper is part of the APL Special Collection on Ferrimagnetic Spintronics.
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ABSTRACT

In this work, we investigate single-shot all-optical switching (AOS) in Tb/Co/Gd/Co/Tb multilayers in an attempt to establish AOS in synthetic ferrimagnets with high perpendicular magnetic anisotropy. In particular, we study the effect of varying Tb thicknesses to disentangle the role of the two rare-earth elements. Even though the role of magnetic compensation has been considered to be crucial, we find that the threshold fluence for switching is largely independent of the Tb content. Moreover, we identify the timescale for the magnetization to cross zero to be approximately within the first ps after laser excitation using time-resolved magneto–optic Kerr effect. We conclude that the switching is governed mostly by interactions between Co and Gd.

The energy efficiency and speed of single-shot helicity-independent all-optical switching (AOS) hold great potential for future memory and storage applications.[1,2] Following the first observation of the phenomenon of AOS in GdFeCo alloys,[3] much progress has been made toward implementing AOS in concrete memory applications. For instance, magnetic tunnel junctions (MTJs) have been shown to be all-optically switchable by replacing the free layer with TM-rare-earth (RE) systems, using Gd-based alloys,[4] Co/Gd multilayers,[5] as well as [Co/Tb] multilayers.[6] Solutions regarding the design of photonic building blocks to address the spintronic devices in an integrated platform environment have also been demonstrated.[7–10]

In all-optically switchable materials, it is deemed essential that the transition metal (TM) sublattice (Fe and Co), and the RE Gd are exchange-coupled antiparallel and exhibit a strong disparity in demagnetization timescales to enable AOS.[1,2,11] However, for high density data storage applications, materials based on the TM-Gd system face significant challenges, largely due to the insufficient perpendicular magnetic anisotropy (PMA) of Gd and the TM system, required to stabilize competitively small domains.

In search for materials with higher magnetic anisotropy, compounds containing the RE element Tb have since gained much attention (see Refs. 12–20). Whereas this improved anisotropy is desirable from a storage density point of view, it was shown to lead to a much faster demagnetization of the 4*f* orbitals compared to Gd. Frietsch *et al.* explained this by a transfer of angular momentum from the 4*f* moments to the magnon and phonon system, which is only possible in the presence of spin–orbit coupling.[21]

While AOS in TM-Gd alloys and multilayers was explained by a strong difference in demagnetization timescales between the TM and the RE—not given for TM-Tb systems—the observation of single-shot AOS in [Co/Tb] multilayers[22,23] came as a surprise. However, the prerequisites for AOS and the exact mechanism behind the magnetization reversal are very different. In multilayers with Tb, AOS is only observed within a small window of compositions close to magnetic compensation, whereas Co/Gd-based multilayers can be engineered to show robust switching even in samples with no magnetic compensation point.[24,25] Another fundamental difference between the two material platforms is the timescale on which reversal occurs. In GdFeCo, it was found that the Fe magnetization crosses zero after only a few hundreds of fs,[26] which can be explained for instance by a phenomenological description,[27–29] exchange scattering,[30] or nonlocal angular momentum transfer.[31] Reversal in [Co/Tb] on the other hand has recently been found to take place on a much longer timescale of ~100 ps,[32] presumably driven by virtue of precessional switching around a transient in-plane (IP) anisotropy field, induced by ultrashort heating.[33] Furthermore, switching in [Co/Tb] was found to be independent of the duration of the laser stimulus in the range between 50 fs and 10 ps (not present in TM-Gd systems[28]) clearly hinting toward slower switching dynamics.[33]

Systems combining both Gd and Tb offer appealing alternatives, using Gd as the main driving force for AOS while exploiting the high magnetic anisotropy of Tb. Recently, AOS was found in systems where Gd makes up at least 20% of the total RE material.[34,35] A downside of alloys on the other hand are limited possibilities of (interfacial) thin film engineering compared to multilayer films.

In this work, we seek to combine the ultrafast and robust switching of Co/Gd bilayers with Tb to disentangle the roles of the two RE elements. To do this, we design multilayer stacks with Co, Gd, and Tb. We study for which RE layer thicknesses the system exhibits PMA and quantify effective anisotropy constants. Furthermore, we address the questions of the dependence of the critical fluence for AOS on magnetic compensation, which has been considered a critical parameter until now. Finally, we investigate the timescales on which this switching occurs.

Samples in this study are deposited by dc magnetron sputter deposition at room temperature on Si/SiO_2 and doped Si:B substrates for static and time-resolved experiments, respectively. For investigating static magnetic properties, the polar magneto–optic Kerr effect (MOKE) is used to record out-of-plane (OOP) hysteresis loops at room temperature. Furthermore, a superconducting quantum interference device vibrating sample magnetometer (SQUID-VSM) is employed to measure IP hysteresis curves at 300 K. Single-shot all-optical switching is triggered by single linearly polarized ~100 fs laser pulses at a central wavelength of 700 nm with variable energy at normal incidence. Optically reversed domains are imaged using Kerr microscopy in polar configuration. For the investigation of time-resolved dynamics, we employ time-resolved MOKE (TR-MOKE) using pump and probe pulses with a pulse duration of <50 fs, central wavelengths of 800 and 400 nm, respectively, and a repetition rate of 5 kHz.

When engineering the synthetic ferrimagnetic sample stack, it is important to consider that the Curie temperatures (T_C) of the bulk Gd and Tb are 292 and 222 K, respectively, and, therefore, below room temperature. Their magnetization will only be stabilized when interfaced with a ferromagnetic material like Co through the magnetic proximity effect, pushing T_C above room temperature. Taking this into account, the choice is made to perform experiments on Ta(4)/Tb(t_{Tb})/Co(0.8)/Gd(t_{Gd})/Co(0.8)/Tb(t_{Tb})/TaN_x(4) multilayer samples (thicknesses in nm). A sketch of the sample design is shown in Fig. 1(a). Comments on the reproducibility of the samples are provided in the supplementary material.

For the first sample, Gd and Tb thicknesses are spatially varied along perpendicular directions to study a wide composition range. In order to determine the composition range where PMA is present, we measure hysteresis loops at different locations along the sample using MOKE. As the MOKE measurement is sensitive to mostly the Co magnetization, a reversal of the sign of the Kerr angle is expected when scanning from Co to RE-dominated regions. We extract the saturation Kerr angle θ_s from our data and show the resulting phase diagram in Fig. 1(b).

Regions with no PMA are shown in white, Co and RE-dominated regions with PMA in blue and red, respectively. We find that PMA is absent in composition regions, where the Tb is thinner than a monolayer (~0.3 nm). We attribute this observation to the onset of a discontinuous Co/Tb interface, which is detrimental to obtaining PMA. In regions with too thin Gd, we explain the lack of OOP anisotropy by the strong IP shape anisotropy of the joint two Co layers, overcoming interfacial anisotropy contributions. In the region with PMA, we observe a transition from Co to RE-dominated behavior. The compensation line separating these regions is indicated by a broken line in Fig. 1(b).

To gain quantitative information on the effective magnetic anisotropy constant K_{eff}, we record IP hysteresis loops using SQUID-VSM. For this purpose, separate samples with a fixed Gd thickness of 1 nm and varying Tb thicknesses are deposited. OOP MOKE and IP SQUID loops are presented in Figs. 2(a) and 2(b), respectively. We

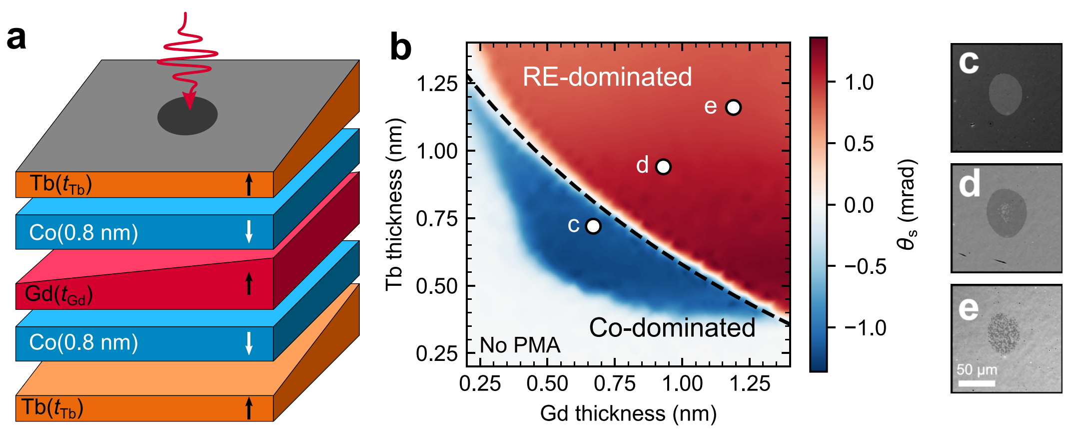

FIG. 1. (a) Sketch showing the magnetic layers within the stack with arrows indicating the local magnetization orientation. Laser excitation occurs with perpendicular incidence from free space. (b) Phase diagram extracted from the saturation Kerr angle of the sample design shown in (a) measured with polar MOKE. The broken line indicates the compensation composition. Co-domination is indicated in blue, RE-domination in red, and IP regions in white. (c)–(e) Kerr microscopy images of the magnetic state of the sample after excitation with a single fs laser pulse at the approximate thicknesses indicated in (b), representing Co with magnetization up (dark) and down (light).

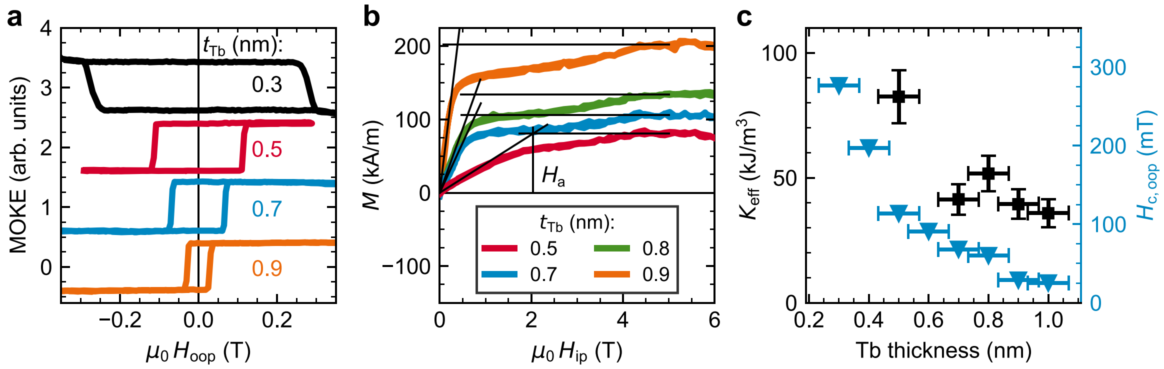

FIG. 2. (a) Perpendicular MOKE hysteresis loops (vertical offsets for clarity) and (b) IP SQUID loops for various Tb thicknesses at a fixed Gd thickness of 1 nm measured at 300 K. A negative MOKE step in a indicates that the sample is Co-dominated. (c) Effective anisotropy and coercive fields extracted from a and b as a function of Tb thickness.

note that the average magnetization in **b** is calculated based on the assumption that the full volume of RE and Co layers contributes to the magnetic moment. Further elaboration on this is provided in the supplementary material. For the Co-dominated sample ($t_{Tb} = 0.3$ nm), the step of the hysteresis loop in Fig. 2(a) is inverted as discussed above. The OOP loops show well-defined square shapes with decreasing coercive fields H_c as t_{Tb} increases [see Fig. 2(c)]. IP loops show typical hard axis reversal at low fields with close to zero remanence and a linear dependence as a function of field, until a plateau is reached. Samples with $t_{Tb} < 0.5$ nm could not be saturated along their hard axis with the available field of 7 T, and corresponding loops are not shown. A peculiarity in all IP measurements is a second slow magnetization step-up to ∼4.5 T. Such behavior has previously been ascribed to the so-called fanning cone in TM-Tb-based samples.[36,37]

To extract the effective uniaxial magnetic anisotropy constant from the IP data, the anisotropy field H_a and saturation magnetization M_s are determined. Due to ambiguities arising from the presence of the second step in the hysteresis loops, the following choices are made for determining M_s and H_a: M_s is assumed to be the value of the magnetization after the second step and H_a is the field at which the line that is fitted to the low-field behavior intersects with M_s. For the $t_{Tb} = 0.5$ nm sample, the extracted lines describing the low-field behavior, a horizontal line at M_s, and a vertical line at H_a are indicated in Fig. 2(b). The effective anisotropy constant is then given by

$$K_{eff} = \frac{1}{2}\mu_0 H_a M_s. \tag{1}$$

Figure 2(c) shows H_c and K_{eff} as a function of t_{Tb}. We observe a steady decrease in K_{eff} as a function of t_{Tb}. This trend could stem from a variety of effects. First, the origin of the anisotropy is interfacial, meaning that it is expected to scale inversely proportional to the total film thickness. Second, adding more Tb could result in a decrease in T_C, which typically leads to a decrease in magnetic anisotropy. Third, a higher RE content drives the material further away from compensation, leading to an increase in shape anisotropy K_s, which decreases PMA. Furthermore, higher thicknesses of Tb may lead to rougher Tb/Co interfaces, which could also contribute to the observed decrease in PMA toward higher Tb thicknesses. Bringing the values of K_{eff} into perspective with literature data on other samples exhibiting AOS, we find anisotropy constants in a similar range as found in (Gd, Tb)Co alloys,[34] yet slightly lower than in Pt/Co(1 nm)/Gd(3 nm) layers.[38,39] We argue that it should be possible to increase K_{eff} through stack engineering, such as by adding more Tb/Co repeats.

We now turn our attention toward the characterization of AOS in our samples. First, we investigate single-shot AOS at different spots in the double wedge sample, indicated in Fig. 1(b). To do so, the sample is first brought to saturation by a perpendicular magnetic field. Subsequently, single fs laser pulses are directed onto the sample surface. We image the irradiated regions using Kerr microscopy and show selected results in Figs. 1(c)–1(e). A more comprehensive set of Kerr images with further discussion is provided in the supplementary material. We find that in **c** and **d**, fully switched magnetic domains are created, which is confirmed by the difference in gray level of the saturated magnetic states. It is possible to switch reversed states back to their initial state upon irradiation with subsequent laser shots. The reversal in contrast from Figs. 1(c) to 1(d) is again caused by a transition from Co to RE dominated regions. However, for higher Gd and Tb thicknesses in Fig. 1(e), only a multi-domain state is present. This is likely due to the fact that in this region, the excess of RE content brings the system too far away from magnetic compensation, preventing AOS. As discussed in Ref. 40, other reasons as to why a multidomain state may arise include the phonon temperature temporarily exceeding T_C, or stray fields facilitating the nucleation of a multidomain state at elevated temperatures. Indeed, a multi-domain state is already visible in Fig. 1(d), although here it only occurs in the middle of the laser pulse, where the fluence is highest.

Having shown that the stack does exhibit AOS, we proceed by investigating the critical fluence for switching and the role of Tb in the process. For this experiment, we again take the sample series with fixed Gd thickness of 1 nm and different Tb thicknesses. After exposing the sample to laser pulses with different energies, the sizes of optically reversed domains are investigated. A typical image is presented in Fig. 3(a) with pulse energies annotated in the figure. The extracted domain size A_{Domain} as a function of pulse energy E is plotted in Fig. 3(b). To quantify the (incident) threshold fluence F_0 for AOS from this measurement, the equation below is used to describe the pulse energy dependence, assuming elliptical laser profiles:[41]

$$A_{Domain} = \pi r \sigma^2 \ln\left(\frac{E}{F_0 \pi r \sigma^2}\right). \tag{2}$$

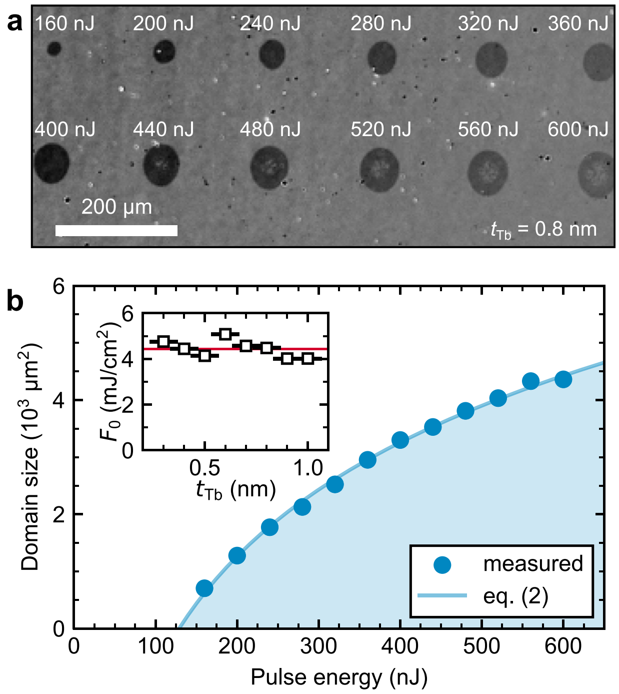

FIG. 3. (a) Kerr microscopy images of the Tb(0.8 nm)/Co/Gd(1 nm)/Co/Tb(0.8 nm) sample after excitation by a single fs linearly polarized laser pulses with energies indicated in the Figure. (b) Sizes of reversed domains as a function of laser pulse energy extracted from (a). The continuous line is a best fit according to Eq. (2). The inset shows the critical fluence for AOS as a function of Tb thickness with the horizontal red line indicating the mean value of 4.44 mJ/cm^2.

σ denotes the length of the ellipse, r is the ratio of short and long axes, and E is the pulse energy. The above equation is fitted to all data sets, and F_0 is extracted, as shown in the inset of Fig. 3(b). We find that F_0 is effectively independent of the Tb thickness with a mean value of 4.44 mJ/cm^2 at the given Gd thickness of 1 nm. This is somewhat surprising, considering the potential for AOS in pure [Co/Tb] multilayers, where AOS is observed only very close to magnetic compensation.[22,23] In the work on TbGdCo alloys, the threshold fluence was found to increase with increasing Tb content,[34] which, however, also entails removal of Gd from the system. We conclude that the switching must originate from the Co/Gd and Gd/Co interfaces, which are known to facilitate ultrafast switching. It is plausible that the Tb moment is fully quenched by the laser due to its low T_C and high spin–orbit coupling and aligns opposite to the switched Co after remagnetization.

We note that the extended switching overview in the supplementary material suggests that for thin Gd, adding larger amounts of Tb even slightly reduces the efficiency of AOS. This notion does not contradict our findings here and could be explained by a deterioration of interface quality.

Finally, we want to address the question of the timescale on which magnetization reversal takes place by measuring the time-resolved response to a fs laser pulse with TR MOKE. To ensure that the magnetization returns to its initial state after each excitation, a variable external OOP field is applied during the experiments. We investigate samples with a Gd thickness of 1 nm and a Tb thickness of 0.3 nm. Time traces for different applied field strengths are shown in Fig. 4(a) at a pump fluence of 5.23 mJ/cm^2, which is greater than F_0. We find that the magnetization crosses zero approximately within the first ps after laser excitation, independently of the magnitude of the field. It is, therefore, not the slow precessional reversal found in [Co/Tb] multilayers, but the fast exchange-driven switching known to exist in Co/Gd systems that facilitates AOS in our samples. A full reversal to the antiparallel state is inhibited by the presence of the guiding field. As the magnetization slowly recovers over the following tens of ps, the magnetic state is reset by virtue of precessional switching, as found in Co/Gd.[42] This process is clearly accelerated by increasing the guiding field, as the measurement recorded in the lowest field of 0.2 T takes the longest time to recover its magnetization.

Furthermore, we study the dynamics at different pump fluences in Fig. 4(b). As the fluence increases, the magnetization is quenched progressively until a zero crossing occurs at a fluence of 4.36 mJ/cm^2.

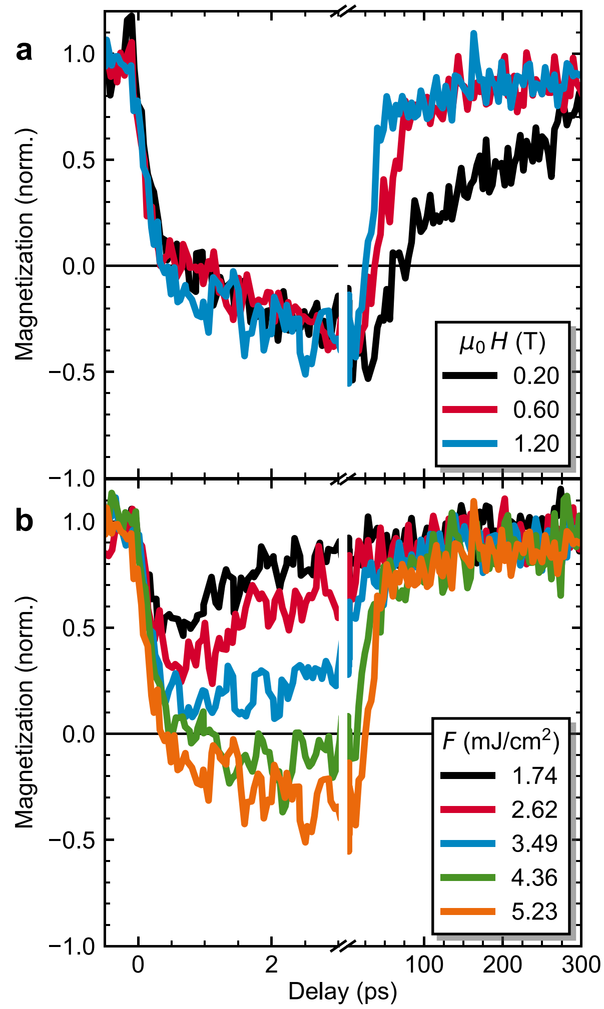

FIG. 4. Time-resolved MOKE measurements of the Tb(0.3 nm)/Co/Gd(1 nm)/Co/Tb(0.3 nm) sample (a) for different applied fields at a fluence of 5.23 mJ/cm^2 and (b) for different laser fluences at an applied field of 1.2 T. The laser excitation occurs at a delay time of 0 ps.

This value agrees reasonably well with $F_0 = 4.44\,\mathrm{mJ/cm^2}$, determined from static experiments. When comparing F_0 with well-established AOS platforms, the values are comparable to those found in pure CoGd alloys,[34] but about twice as high as those in Pt/Co(1 nm)/Gd(3 nm) multilayers.[41] This discrepancy could be attributed to a higher ratio of Co:Gd due to the different stack architecture.

In summary, we found PMA and robust and ultrafast AOS in Tb/Co/Gd/Co/Tb samples in a wide composition range. The zero crossing of the magnetization occurred approximately within the first ps after laser excitation, leading to the conclusion that the mechanism for reversal is identical as in Co/Gd. The fact that the addition of Tb does not result in a reduction of the critical fluence supports this notion. Our results show that hybrid structures based on Co/Gd and Co/Tb multilayers are promising candidates for magneto-photonic memory devices.

See the supplementary material for further details about sample fabrication, a more comprehensive overview on AOS on the double wedge sample, and additional information on the magnetostatic characterization.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie Grant Agreement Nos. 861300 and 860060. This work was also part of the research program Gravitation (ZK) program "Research Center for Integrated Nanophotonics," which are financed by the Dutch Research Council (NWO). J.I. acknowledges support from JSPS Overseas Research Fellowships. Furthermore, this work was supported by the No. ANR-20-CE09-0013 UFO, the Institute Carnot ICEEL for the project "CAPMAT" and FASTNESS, the Région Grand Est, the Metropole Grand Nancy, for the Chaire PLUS by the impact project LUE-N4S, part of the French PIA project "Lorraine Université d'Excellence" Ref. No. ANR-15-IDEX-04-LUE, the "FEDERFSE Lorraine et Massif Vosges 2014–2020" for IOMA a European Union Program, and the ANR Project No. ANR-20-CE24-0003 SPOTZ.

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Julian Hintermayr: Conceptualization (equal); Investigation (lead); Methodology (equal); Writing – original draft (lead); Writing – review & editing (lead). **Pingzhi Li:** Investigation (equal); Methodology (equal); Writing – original draft (supporting); Writing – review & editing (supporting). **Roy Rosenkamp:** Conceptualization (equal); Investigation (equal); Methodology (equal); Writing – original draft (supporting); Writing – review & editing (supporting). **Youri L. W. Van Hees:** Conceptualization (equal); Investigation (equal); Methodology (equal); Writing – original draft (supporting); Writing – review & editing (supporting). **Junta Igarashi:** Funding acquisition (equal); Methodology (supporting); Writing – original draft (supporting); Writing – review & editing (supporting). **Stéphane Mangin:** Funding acquisition (equal); Investigation (supporting); Resources (equal); Supervision (supporting); Writing – original draft (supporting); Writing – review & editing (supporting). **Reinoud Lavrijsen:** Funding acquisition (equal); Investigation (supporting); Resources (equal); Supervision (supporting); Writing – original draft (supporting); Writing – review & editing (supporting). **Gregory Malinowski:** Funding acquisition (equal); Investigation (supporting); Methodology (supporting); Resources (equal); Supervision (supporting); Validation (equal); Writing – original draft (supporting); Writing – review & editing (supporting). **Bert Koopmans:** Conceptualization (equal); Funding acquisition (equal); Investigation (supporting); Resources (equal); Supervision (lead); Validation (equal); Writing – original draft (supporting); Writing – review & editing (supporting).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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