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1 **From toggle to precessional single laser pulse switching**

2

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12 **Keywords:**

13 Thin films, Rare Earth, Fast optics, Single pulse reversal, Spintronics

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16

17 **Abstract**

18

19 With the advent of nanotechnologies, it has been possible to extend the number of stimuli that
20 can be used to control the state of a magnetic nanostructure. Among those stimuli, single laser
21 pulse excitation allows, under certain conditions, to obtain energy efficient ultra-fast
22 magnetization reversal. With this respect, two different types of single pulse switching
23 mechanisms have been reported. The first one consists in a sub picosecond ultrafast toggle
24 switching which was observed mainly in Gd based alloys. The second type relies on sub-
25 nanosecond precessional switching occurring in Rare Earth–Transition Metal
26 alloys/multilayers. Here, we demonstrate that single pulse all optical switching is achieved in
27 $\text{Co}_{68}\text{Tb}_{32}/\text{Co}_{100-x}\text{Gd}_x/\text{Co}_{68}\text{Tb}_{32}$ tri-layers in which the behavior can be tuned from toggle to
28 precessional by changing the composition of the $\text{Co}_{100-x}\text{Gd}_x$ alloy.

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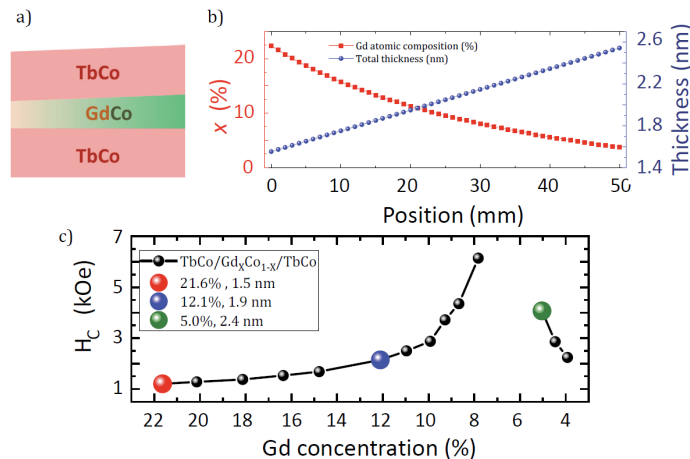
31 The challenge of controlling the magnetization at picosecond timescales in an energy efficient
32 way is at the heart of future magnetic storage devices. With the advent of nanotechnologies, it
33 has been possible to extend the number of stimuli that can be used to control the magnetic state
34 of a magnetic material. In the form of thin films, multilayers or nanostructures, in a non-
35 exhaustive list, the effects of strains, temperature gradients and spin-polarised currents are
36 exasperated, allowing the magnetisation to be reversed without any applied magnetic field.
37 On the other hand, the advent of femtosecond lasers has allowed the exploration of the
38 magnetization dynamics on the ultrafast timescale¹. Furthermore, it has been shown that, under
39 certain conditions, ultrafast magnetization reversal can be achieved in a very energy-efficient
40 process by the use of a single laser pulse². Therefore, this research field area has caught the
41 attention of a large part of the magnetism community. Those developments follow the major
42 discovery of Bigot *et al.* in 1996 that a femtosecond laser pulse can demagnetize at a sub-
43 picosecond time scale a Ni thin film¹. Ten years later Stanciu *et al.* showed deterministic
44 switching of a GdFeCo ferrimagnetic magnetic moment using a single circularly-polarized
45 laser pulse² and later on, a toggle switching independent of the light helicity^{3,4}. The current
46 understanding is that the strong nonequilibrium electronic state and the transfer of angular
47 momentum by exchange or spin current between antiferromagnetically exchanged coupled
48 subnetworks are responsible for the toggle switching in Gd-based materials such as GdFeCo
49 alloys⁴, Gd/Co multilayers⁵ and GdTbCo⁶. The same arguments hold for Mn₂Ru_xGa Heusler
50 ferrimagnetic alloys which possess two inequivalent Mn sublattices and where ultrafast all-
51 optical switching has been observed⁷. All other tests with rare earth alloys have failed so far.
52 Interestingly, single pulse precessional switching has been shown in Tb/Co multilayer^{8,9,10} and
53 extended in a large variety of magnetic multilayers containing different rare earth (RE) (Tb,
54 Dy) and transition metals (TM) (Co, Py and Fe) elements¹¹. Taking advantage of ultrafast heat
55 transfer to the lattice in RE-TM alloys, the in-plane reorientation of the magnetization of the
56 multilayer launches a precessional reversal. In this process, the reversal threshold is
57 independent of the pulse duration for pulse duration lower than 10ps. Here, we demonstrate
58 that single pulse switching is achieved in Co₆₈Tb₃₂/Co_{100-x}Gd_x/Co₆₈Tb₃₂ tri-layers whose
59 behavior can be tuned from toggle to precessional by changing the Co_{100-x}Gd_x concentration.

60
61 The multilayers were grown by sputtering in an AJA system with a base pressure less than
62 5×10^{-8} mbar to ensure oxygen free deposition of RE materials. The sample is made of
63 Glass/Ta(5 nm)/Pt(5 nm)/Co₆₈Tb₃₂(4 nm)/Co_{100-x}Gd_x(t)/Co₆₈Tb₃₂(4 nm)/Ta(5 nm)/Pt(1 nm) as
64 shown in figure 1(a). The Ta/Pt buffer and capping layers ensure adhesion of the substrate and
65 protection against oxidation. The alloys are made by co-deposition of different sources
66 arranged in a confocal geometry, the composition being adjusted by the powers applied on the
67 different guns. For all layers, except for the Co_{100-x}Gd_x(t) layer, the homogeneity of the layer is
68 obtained by rotating the sample. The Co_{100-x}Gd_x(t) is obtained by co sputtering of Co and Gd
69 pure targets. Composition/thickness wedge is obtained by stopping the rotation of the sample
70 during deposition. Since Co and Gd are mounted diametrically opposed, a natural gradient of
71 concentration is obtained. Since the deposition rate of Co and Gd are not identical, a gradient
72 of thickness also exists. A calibration of the position dependent deposition rate has been done¹¹.
73 The local thickness and composition could be calculated. The values are reported in figure 1(b).
74 In full films, Co_{100-x}Gd_x is known to have its compensation around $x = 20\%$ ^{12,13}. One must be
75 aware that the Co_{100-x}Gd_x interfaces can affect the compensation composition as it was shown
76 in ref [14] especially in the case of thin layers. For thicknesses less than 4 nm, it was shown
77 that Co_{100-x}Gd_x can have a perpendicular to the film plane magnetic anisotropy (PMA)¹⁵.
78 In the multilayer, the Co₆₈Tb₃₂(4 nm) are kept constant. Considering the concentration,
79 Co₆₈Tb₃₂ is Tb dominant. Due to the high RE content, the alloy has a Curie temperature roughly
80 373 K above room temperature¹² and possesses PMA.

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84 **Figure 1.** (a) Schematic of the trilayer stack; (b) Variation of the thickness (blue) and Gd
 85 composition (red) as a function of position in the wedge sample; (c) Variation of coercive field,
 86 H_c , versus Gd composition. Red, blue and green dots correspond to the composition of the
 87 samples whose reversal is shown in figure 2.
 88

89 In $\text{Co}_{68}\text{Tb}_{32}(4 \text{ nm})/\text{Co}_{100-x}\text{Gd}_x(t)/\text{Co}_{68}\text{Tb}_{32}(4 \text{ nm})$, the Co atoms are strongly ferromagnetically
 90 exchange coupled at the interface. Therefore, Tb and Gd magnetization subnetwork are
 91 parallel. We performed Kerr magnetometer hysteresis loops in the polar geometry to check
 92 PMA, coercivity and subnetwork dominance. As can be seen in figure 1(c), a divergence of the
 93 coercive field associated with the change of subnetwork dominance is the sign of the net
 94 magnetisation compensation of the $\text{Co}_{68}\text{Tb}_{32}(4 \text{ nm})/\text{Co}_{100-x}\text{Gd}_x(t)/\text{Co}_{68}\text{Tb}_{32}(4 \text{ nm})$ trilayer. It
 95 appears with x around 6 to 7 % confirming the Tb dominance in the $\text{Co}_{68}\text{Tb}_{32}$ alloy (more Co
 96 in $\text{Co}_{100-x}\text{Gd}_x$ is needed to get the total compensation). In all cases, PMA could be observed.

97 For the single shot reversal experiments, a Ti:sapphire femtosecond-laser source was used with
 98 a wavelength of 800 nm and a 5 kHz repetition rate for the pump-probe experiments. LED light
 99 probe source with a wavelength of 628 nm was used to obtain MOKE images. Since samples
 100 have been grown on glass substrate, pump laser excitation was done on the glass side of the
 101 sample while MOKE microscope observation was done on the layers side. Details on the
 102 sample growth, on MOKE microscope and pump-probe setups can be found in [11].

103 Single pulse magnetization switching experiments have been done for concentration range x
 104 from 5 to 22 % and measurements performed for $x = 5 \%$, 12.1 % and 21.6 % are reported in
 105 figure 2a. When $x = 21.6 \%$, a perfect single shot switching is observed. On the other hand,
 106 when $x = 5 \%$, single-shot switching was observed along with the stabilization of a ring
 107 structure. In between, a multi-domain state occurs as seen for $x = 12.1 \%$. A more detailed
 108 study of reversal mechanism is obtained by measuring state diagrams that report the static
 109 magnetic state as a function of both pulse duration and fluence as shown in figure 2 b and c.

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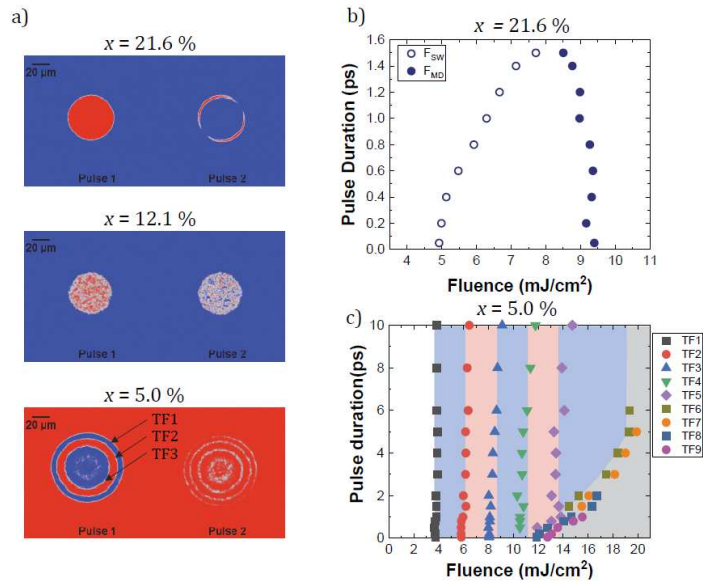


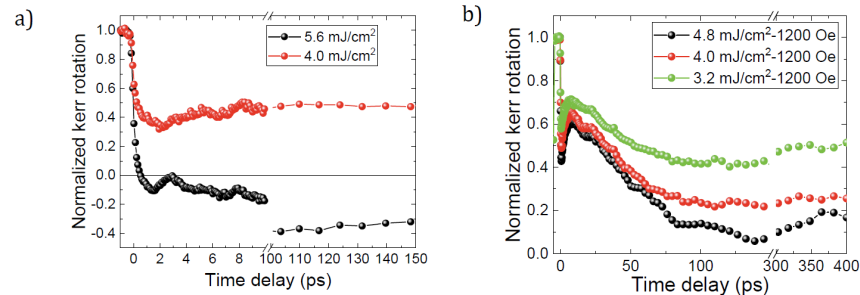
Figure 2. (a) Single pulse reversal experiments done for the samples with 21.6 % (5.9 mJ/cm^2), 12.1 % (10.3 mJ/cm^2) and 5 % (10.3 mJ/cm^2) Gd contents, identified in red, blue and green in figure 1c. Pulse duration versus fluence state diagram measured for 21.6 % of Gd (b) and 5 % of Gd (c). In b) TF_{SW} and TF_{MD} correspond to the fluence to get single shot switching or multidomain state respectively. In c) the complex domain pattern require the definition of multiple threshold fluence : TF_1 corresponds to the appearance of outer blue ring and can be referred as F_{SW} for the ring structure, TF_2 to the appearance of the red ring etc... TF_9 corresponds to TF_{MD} .

The state diagrams differ strongly when $x = 21.6\%$ and $x = 5\%$. As far as the state diagram of $x = 21.6\%$ is concerned (figure 2b), two fluence thresholds can be defined : F_{SW} and F_{MD} that correspond to the fluence needed to induce the switching and a multidomain state respectively. The triangular shape of this state diagram is characteristic of toggle reversal in Gd-based materials¹⁶. Since F_{SW} depends on pulse duration, the underlying mechanism is likely governed by the electronic temperature which decreases with increasing the pulse duration as discussed in [16,17]. On the other hand, since F_{MD} depends only slightly on pulse duration, a link to the Curie temperature, the energy transferred to the lattice and so the stabilisation of multi-domain structure is highly probable as suggested in [17,18,19]. As far as the state diagram of $x = 5\%$ is concerned, the domain pattern is much more complex and multiple threshold fluence have to be defined TF_i ¹¹ : TF_1 corresponds to the appearance of outer blue ring and can be referred as F_{SW} for the ring structure, TF_2 to the appearance of the red ring etc... The fact that the critical fluences (TF_1 , TF_2 ,...) are mostly independent of pulse duration implies an underlying mechanism linked to the energy transferred to the lattice¹¹. A careful analysis of the position of the boundaries that separates the rings has shown that they are always located at the same local threshold energy¹¹, independently of surrounding domains and/or local in-plane heat gradients.

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140 More insight in the reversal process is gained by the analysis of the pump-probe measurements.
 141 Results are reported in figure 3. For the sample with $x = 21.6\%$, dynamics of reversal has been
 142 measured for two fluences : $F = 4.0 \text{ mJ/cm}^2$ and $F = 5.6 \text{ mJ/cm}^2$ that corresponds to fluences
 143 for which reversal does not occur and occurs respectively when pulse duration is fixed to 50 fs
 144 (see figure 2b). We can clearly see in figure 3a that typical demagnetization occurs and
 145 magnetization crosses zero at 600 fs. Full reversal occurs at 40 ps before magnetization
 146 recovers at longer time scale because of applied field to reset the magnetization for the pump
 147 probe experiments. As far as the sample with $x = 5\%$ is concerned, the measurements show
 148 that after a fast demagnetization at a time scale of some 100 fs, the magnetization first recover,
 149 similarly to the conventional demagnetization/re-magnetization shown in various
 150 ferromagnetic and ferrimagnetic materials. At longer timescales, a few tens of picoseconds, a
 151 magnetization decrease is observed. This decrease is attributed to the reorientation of the
 152 magnetization in the plane of the layers¹¹. The existence of the DC field, in combination to a
 153 strongly damped reorientation of the anisotropy, makes the precession difficult to be
 154 observed¹¹.
 155



156
 157 **Figure 3.** Pump-probe dynamics measured for 21.6 % of Gd (a) and 5 % of Gd (b) at different
 158 fluences. The pulse duration has been fixed to 50 fs.
 159
 160

161 A summary of the different fluence threshold measured as a function of Gd content and for a
 162 pulse duration fixed to 50 fs is reported in figure 4.
 163

164 We can clearly see that for x above 12.5 %, the $\text{Co}_{68}\text{Tb}_{32}(4 \text{ nm})/\text{Co}_x\text{Gd}_{100-x}(\text{t})/\text{Co}_{68}\text{Tb}_{32}(4 \text{ nm})$
 165 trilayer behaves like Gd based materials presenting single shot ultrafast toggle switching: 1)
 166 F_{sw} increases as the Gd content decreases within the range of 22 % to 16 %. The deterministic
 167 switching of the CoGd alloy is conditioned by the amount of spin current generated through
 168 the demagnetization of the Gd subnetwork. The increase of F_{sw} indicates that the
 169 demagnetization of Gd needs to be increased to keep a significant spin current since its
 170 concentration is decreased. 2) Conversely, as the magnetic compensation point occurs around
 171 $x = 20\%$ [12,13], the saturation magnetization of CoGd (and demagnetizing fields) increases
 172 as the Gd content decreases from 16 % to 12 %. This increase in saturation magnetization,
 173 coupled with a decrease in anisotropy, results in an overall reduction in effective anisotropy,
 174 promoting the formation of magnetic domains. These changes in the 16-to-12 % range lead to
 175 a reduction of the critical threshold for multi-domain formation.
 176

177 In the 8-to-12.5 % range, F_{MD} becomes smaller than F_{sw} , and therefore only a final
 178 multidomain state is observed. Interestingly, in this range of composition, a close analysis of

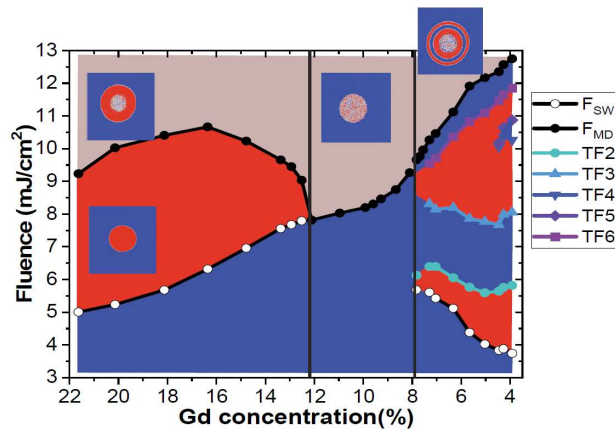
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179 the magneto-optical signal (figure 2a, 12.1 %) reveals that a fraction of the magnetization
 180 toggles after each laser shot, even though the majority of the exposed area appears
 181 demagnetized. Possibly, a transient reversal might be still taking place in the 8-to-12.5 % range,
 182 which is then hindered or erased by the demagnetizing fields that promote multi-domain
 183 formation.

184
 185 Finally, when x is less than 8 %, the $\text{Co}_{68}\text{Tb}_{32}(4 \text{ nm})/\text{Co}_x\text{Gd}_{100-x}(t)/\text{Co}_{68}\text{Tb}_{32}(4 \text{ nm})$ undergoes
 186 a precessional reversal with the appearance of multiple threshold fluences¹¹. As can be seen in
 187 figure 4, it occurs even for rather low fluences (down to 3.5 mJ/cm^2). This behaviour is
 188 observed when the $\text{Co}_x\text{Gd}_{100-x}$ layer undergoes an in-plane reorientation that launches the
 189 precession of magnetization¹¹. Finally, we believe that another key ingredient for the reversal
 190 around this composition range is the rather low overall saturation magnetization (and
 191 demagnetizing fields) of the stacks which increases the multi-domain threshold and stabilizes
 192 large domains.

193
 194 In order to cover the composition range from 4 % et 22 % of Gd, in the geometry of our AJA
 195 PVD system, different deposition rates have to be used for Gd and Co. This leads unfortunately
 196 also to a wedge in thickness. We know from our previous work that the thickness of the TM
 197 layer in RE-TM alloy/TM/RE-TM alloy trilayers has an impact on the precessional reversal
 198 [11]. Therefore, we tested if the thickness t of $\text{Co}_x\text{Gd}_{100-x}(t)$, with t around the thickness of
 199 $\text{Co}_{95}\text{Gd}_5(2.6 \text{ nm})$, has an impact on the reversal in the 20 % to 10 % composition range. We
 200 found that the toggle switching is kept for t up to 3.5 nm. Only the size of the reversed domain
 201 and the composition to get into demagnetized state slightly change. So, we undoubtedly
 202 demonstrate that the concentration x is the most important parameter to consider in this study.
 203
 204



205
 206 **Figure 4.** Threshold fluences as a function of Gd concentration for a pulse duration fixed to 50
 207 fs. The colors correspond to the magnetic state starting from a saturated blue state. At low
 208 fluences, the state is not modified. At high fluence, the pink area denotes a demagnetized state.
 209 In between, the behavior depends on gadolinium concentration.
 210

211 In conclusion, we demonstrate that single pulse all optical switching is achieved in
 212 $\text{Co}_{68}\text{Tb}_{32}/\text{Co}_{100-x}\text{Gd}_x/\text{Co}_{68}\text{Tb}_{32}$ tri-layers in which the behavior can be tuned from toggle to

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213 precessional by changing the composition of the $\text{Co}_{100-x}\text{Gd}_x$ alloy. In order to promote toggle
214 switching, $\text{Co}_{100-x}\text{Gd}_x$ alloy needs to keep PMA. Furthermore, the spin current coming from the
215 demagnetization of Gd has to be large enough and remains the key ingredient to promote the
216 toggle switching. This behavior stands till a multidomain state is observed which is a sign that
217 $F_{\text{SW}} > F_{\text{MD}}$. On the other hand, precessional switching occurs for low Gd concentration. In this
218 case, the spin current generated by the Gd demagnetization is strongly decreased while the net
219 magnetization increases and so does the demagnetization field, favoring the in-plane
220 reorientation of the magnetization. This work emphasize the role of magnetic anisotropy on the
221 laser induced ultrafast magnetization reversal in complex magnetic multilayers which is
222 foreseen as a candidate for magneto-photonic memory applications.
223

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