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Abstract

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

The challenge of controlling the magnetization at picosecond timescales in an energy efficient way is at the heart of future magnetic storage devices. With the advent of nanotechnologies, it has been possible to extend the number of stimuli that can be used to control the magnetic state of a magnetic material. In the form of thin films, multilayers or nanostructures, in a non-exhaustive list, the effects of strains, temperature gradients and spin-polarised currents are exhasperated, allowing the magnetisation to be reversed without any applied magnetic field. On the other hand, the advent of femtosecond lasers has allowed the exploration of the magnetization dynamics on the ultrafast timescale[1]. Furthermore, it has been shown that, under certain conditions, ultrafast magnetization reversal can be achieved in a very energy-efficient process by the use of a single laser pulse[2]. Therefore, this research field area has caught the attention of a large part of the magnetism community. Those developments follow the major discovery of Bigot *et al. in 1996* that a femtosecond laser pulse can demagnetize at a sub-picosecond time scale a Ni thin film[1]. Ten years later Stanciu et al. showed deterministic switching of a GdFeCo ferrimagnetic magnetic moment using a single circularly-polarized laser pulse[2] and later on, a toggle switching independent of the light helicity[3,4]. The current understanding is that the strong nonequilibrium electronic state and the transfer of angular momentum by exchange or spin current between antiferromagnetically exchanged coupled subnetworks are responsible for the toggle switching in Gd-based materials such as GdFeCo alloys[4], Gd/Co multilayers[5] and GdTbCo[6]. The same arguments hold for Mn_2Ru_xGa Heusler ferrimagnetic alloys which possess two inequivalent Mn sublattices and where ultrafast all-optical switching has been observed[7]. All other tests with rare earth alloys have failed so far. Interestingly, single pulse precessional switching has been shown in Tb/Co multilayer[8,9,10] and extended in a large variety of magnetic multilayers containing different rare earth (RE) (Tb, Dy) and transition metals (TM) (Co, Py and Fe) elements[11]. Taking advantage of ultrafast heat transfer to the lattice in RE-TM alloys, the in-plane reorientation of the magnetization of the multilayer launches a precessional reversal. In this process, the reversal threshold is independent of the pulse duration for pulse duration lower than 10ps. Here, we demonstrate that single pulse switching is achieved in $Co_{68}Tb_{32}/Co_{100-x}Gd_x/Co_{68}Tb_{32}$ tri-layers whose behavior can be tuned from toggle to precessional by changing the $Co_{100-x}Gd_x$ concentration.

The multilayers were grown by sputtering in an AJA system with a base pressure less than 5×10^{-8} mbar to ensure oxygen free deposition of RE materials. The sample is made of Glass/Ta(5 nm)/Pt(5 nm)/$Co_{68}Tb_{32}$(4 nm)/$Co_{100-x}Gd_x$(t)/$Co_{68}Tb_{32}$(4 nm)/Ta(5 nm)/Pt(1 nm) as shown in figure 1(a). The Ta/Pt buffer and capping layers ensure adhesion of the substrate and protection against oxidation. The alloys are made by co-deposition of different sources arranged in a confocal geometry, the composition being adjusted by the powers applied on the different guns. For all layers, except for the $Co_{100-x}Gd_x$(t) layer, the homogeneity of the layer is obtained by rotating the sample. The $Co_{100-x}Gd_x$(t) is obtained by co sputtering of Co and Gd pure targets. Composition/thickness wedge is obtained by stopping the rotation of the sample during deposition. Since Co and Gd are mounted diametrically opposed, a natural gradient of concentration is obtained. Since the deposition rate of Co and Gd are not identical, a gradient of thickness also exists. A calibration of the position dependent deposition rate has been done[11]. The local thickness and composition could be calculated. The values are reported in figure 1(b). In full films, $Co_{100-x}Gd_x$ is known to have its compensation around x = 20 %[12,13]. One must be aware that the $Co_{100-x}Gd_x$ interfaces can affect the compensation composition as its was shown in ref [14] especially in the case of thin layers. For thicknesses less than 4 nm, it was shown that $Co_{100-x}Gd_x$ can have a perpendicular to the film plane magnetic anisotropy (PMA)[15].
In the multilayer, the $Co_{68}Tb_{32}$(4 nm) are kept constant. Considering the concentration, $Co_{68}Tb_{32}$ is Tb dominant. Due to the high RE content, the alloy has a Curie temperature roughly 373 K above room temperature[12] and possesses PMA.

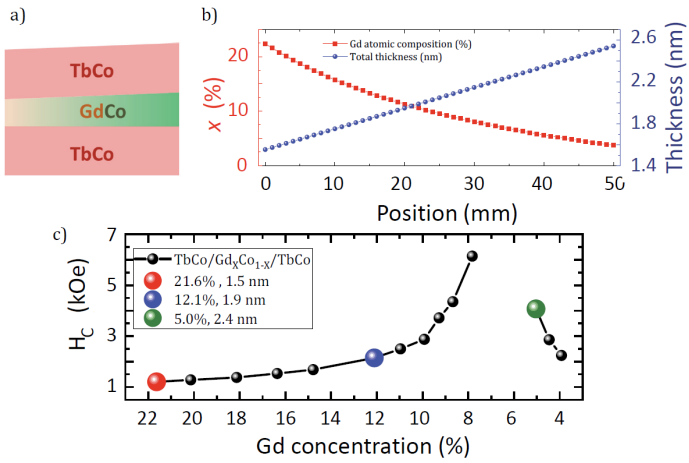

Figure 1. (a) Schematic of the trilayer stack; (b) Variation of the thickness (blue) and Gd composition (red) as a function of position in the wedge sample; (c) Variation of coercive field, H_C, versus Gd composition. Red, blue and green dots correspond to the composition of the samples whose reversal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

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

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

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

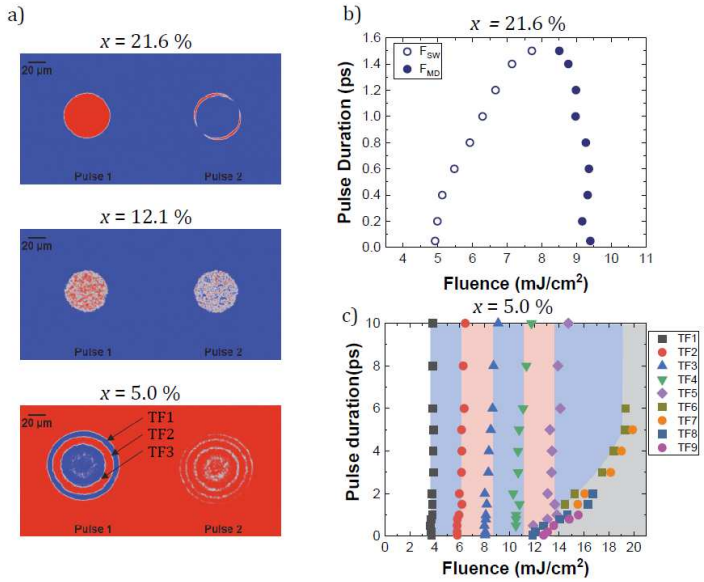

Figure 2. (a) Single pulse reversal experiments done for the samples with 21.6 % (5.9 mJ/cm^2), 12.1 % (10.3 mJ/cm^2) and 5 % (10.3 mJ/cm^2) Gd contents, identified in red, blue and green in figure 1c. Pulse duration versus fluence state diagram measured for 21.6 % of Gd (b) and 5 % of Gd (c). In b) 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 : TF1 corresponds to the appearance of outer blue ring and can be referred as F_{SW} for the ring structure, TF2 to the appearance of the red ring etc…TF9 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 materials[16]. 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_n[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 lattice[11]. 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 energy[11], independently of surrounding domains and/or local in-plane heat gradients.

More insight in the reversal process is gained by the analysis of the pump-probe measurements. Results are reported in figure 3. For the sample with $x = 21.6$ %, dynamics of reversal has been measured for two fluences : F = 4.0 mJ/cm^2 and F = 5.6 mJ/cm^2 that corresponds to fluences for which reversal does not occur and occurs respectively when pulse duration is fixed to 50 fs (see figure 2b). We can clearly see in figure 3a that typical demagnetization occurs and magnetization crosses zero at 600 fs. Full reversal occurs at 40 ps before magnetization recovers at longer time scale because of applied field to reset the magnetization for the pump probe experiments. As far as the sample with $x = 5$ % is concerned, the measurements show that after a fast demagnetization at a time scale of some 100 fs, the magnetization first recover, similarly to the conventional demagnetization/re-magnetization shown in various ferromagnetic and ferrimagnetic materials. At longer timescales, a few tens of picoseconds, a magnetization decrease is observed. This decrease is attributed to the reorientation of the magnetization in the plane of the layers[11]. The existence of the DC field, in combination to a strongly damped reorientation of the anisotropy, makes the precession difficult to be observed[11].

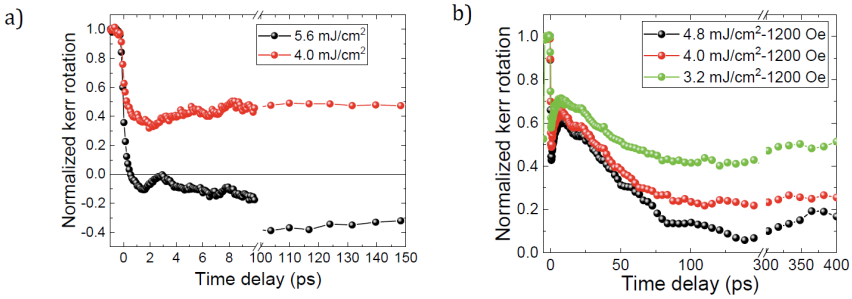

Figure 3. Pump-probe dynamics measured for 21.6 % of Gd (a) and 5 % of Gd (b) at different fluences. The pulse duration has been fixed to 50 fs.

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

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

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

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

Finally, when x is less than 8 %, the $Co_{68}Tb_{32}$(4 nm)/Co_xGd_{100-x}(t)/$Co_{68}Tb_{32}$(4 nm) undergoes a precessional reversal with the appearance of multiple threshold fluences[11]. As can be seen in figure 4, it occurs even for rather low fluences (down to 3.5 mJ/cm^2). This behaviour is observed when the Co_xGd_{100-x} layer undergoes an in-plane reorientation that launches the precession of magnetization[11]. Finally, we believe that another key ingredient for the reversal around this composition range is the rather low overall saturation magnetization (and demagnetizing fields) of the stacks which increases the multi-domain threshold and stabilizes large domains.

In order to cover the composition range from 4 % et 22 % of Gd, in the geometry of our AJA PVD system, different deposition rates have to be used for Gd and Co. This leads unfortunately also to a wedge in thickness. We know from our previous work that the thickness of the TM layer in RE-TMalloy/TM/RE-TMalloy trilayers has an impact on the precessional reversal [11]. Therefore, we tested if the thickness t of $Co_xGd_{100-x}(t)$, with t around the thickness of $Co_{95}Gd_5$(2.6 nm), has an impact on the reversal in the 20 % to 10 % composition range. We found that the toggle switching is kept for t up to 3.5 nm. Only the size of the reversed domain and the composition to get into demagnetized state slightly change. So, we undoubtedly demonstrate that the concentration x is the most important parameter to consider in this study.

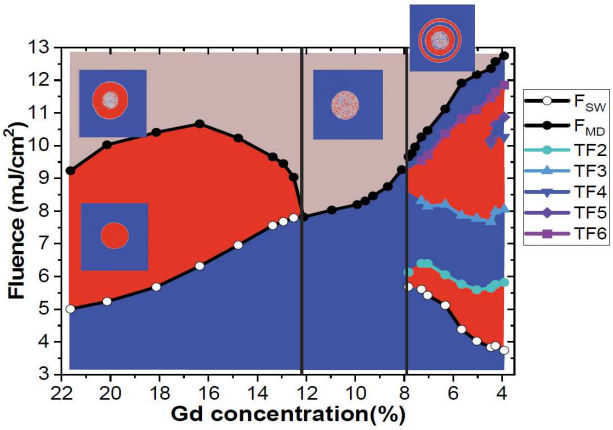

Figure 4. Threshold fluences as a function of Gd concentration for a pulse duration fixed to 50 fs. The colors correspond to the magnetic state starting from a saturated blue state. At low fluences, the state is not modified. At high fluence, the pink area denotes a demagnetized state. In between, the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

In conclusion, we demonstrate that single pulse all optical switching is achieved in $Co_{68}Tb_{32}/Co_{100-x}Gd_x/Co_{68}Tb_{32}$ tri-layers in which the behavior can be tuned from toggle to

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

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