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## Ultrafast Single Pulse Switching of Tb-dominant CoTb Alloy

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Single-pulse magnetization switching by femtosecond laser pulses is the fastest way to manipulate magnetization. To date, among rare-earth transition metal alloys, the single pulse switching is limited to Gd-based structures. Here, we demonstrate the ultrafast single pulse switching of Tb-dominant CoTb alloy within several tens of picoseconds. Our further analysis shows that the ultrafast magnetization reversal is linked to the ultrafast heating of laser pulses and an external field.

Ultrafast manipulation of magnetization is vital for the development of high-frequency spintronic devices. The ultrafast demagnetization in Ni was the first successful example of magnetization control at sub-picosecond timescale <sup>1</sup>. Later, it was reported that the magnetization of GdFeCo alloy reverses back and forth upon the excitation of femtosecond-laser pulses without an external field <sup>2</sup>. This phenomenon is referred to as all-optical helicity-independent switching (AO-HIS) <sup>3</sup>. The picosecond reversal of GdFeCo allows spintronic devices to operate at a repetition rate of up to 3 GHz <sup>4</sup>.

The mechanism of all-optical switching in ferrimagnetic GdFeCo has been studied extensively in the past decade <sup>5</sup>. GdFeCo belongs to rare-earth transition metal (RE-TM) ferrimagnets containing two antiferromagnetically coupled sublattices <sup>6</sup>. In addition, RE and TM magnetic sublattices show different temperature dependencies <sup>7</sup>. Therefore, the magnetization compensation temperature  $T_M$  occurs when the net

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magnetization cancels out. Stanciu *et al.* showed that the femtosecond heating of the GdFeCo alloy with  $T_M$  higher than room temperature leads to subpicosecond reversal<sup>8</sup>. Radu *et al.* showed that the difference in reversal time between Gd and Fe after laser heating leads to a transient ferromagnetic state, which triggers the single pulse ultrafast switching of GdFeCo<sup>2,9</sup>. Recent reports analyzed the role of laser duration, composition, thickness, and damping on AO-HIS<sup>10-13</sup>. Theoretical and experimental results show that the pulse duration required for switching is as long as 15 ps in GdFeCo samples with low damping<sup>14</sup>.

Lalieu *et al.* showed that single-pulse AO-HIS could also be accomplished in Gd-based ferrimagnetic multilayers<sup>15</sup>. However, Beens *et al.* showed that the interfacial mixing between Gd and Co at the interface is critical for single pulse AO-HIS<sup>16</sup>. Our previous study showed that the Tb doping above 5% in GdFeCo eliminates the AO-HIS<sup>17</sup>. Recently AO-HIS has also been observed in MnRuGa, which has two different antiparallel Mn sublattices<sup>18,19</sup>. Magnetic tunnel junctions consisting of Tb/Co nanolayers<sup>20</sup> and Pt/Co/Pt stack<sup>21</sup> have also been reported for AO-HIS.

For RE-TM alloy, the single pulse switching is generally only observed in GdFeCo<sup>22,23</sup>. This paper explores the feasibility of single pulse switching in Gd-free RE-TM, i.e., CoTb alloy. CoTb alloys exhibit strong perpendicular magnetic anisotropies<sup>7,22,23</sup>, which makes them attractive for applications. Recent reports indicated that partial single-pulse switching could be performed in CoTb alloys<sup>24-26</sup>, suggesting that it has the potential to achieve complete single-pulse switching.

To date, evidence of single pulse switching in non-Gd-based RE-TM alloy is still lacking. In this study, we witnessed single pulse switching in Tb-rich CoTb alloy. Our results indicate that the single pulse switching is stimulated by femtosecond laser pulses and an opposite external field.

We explored the ultrafast magnetization dynamics of CoTb upon femtosecond laser excitation by time-resolved Magneto-optical Kerr effect (TRMOKE), as shown in Fig. 1(a). The TRMOKE system was built using an amplified Ti: Sapphire laser system with 35 fs laser pulses at a central wavelength of 800 nm and a repetition rate of 1 kHz. The laser is divided into the pump beam and the probe beam. The probe beam with a wavelength of 400 nm is used to probe the Co sublattice. The fluence of the probe beam is far less than that of the pump beam to eliminate the thermal effect of the probe laser. The pump beam is linearly polarized to eliminate the influence of helicity. We prepared Ta (5 nm)/Co<sub>0.76</sub>Tb<sub>0.24</sub> (6 nm)/Ta (2 nm) thin film of 10×10 mm<sup>2</sup> by magnetron sputtering, while Ta (2 nm) is the capping layer (the inset in Fig. 1(b)). Fig. 1(b) shows the ultrafast process of

$\text{Co}_{0.76}\text{Tb}_{0.24}$  alloy with an external magnetic field  $H_{\text{ext}}$  of 15 kOe. After full demagnetization, the magnetization of the Co sublattice reversed to the opposite magnetization state. Previous studies have not reported the switching dynamic of CoTb alloy triggered by a single pulse. Our results support that single pulse switching also applies to non-Gd-based RE-TM alloy.

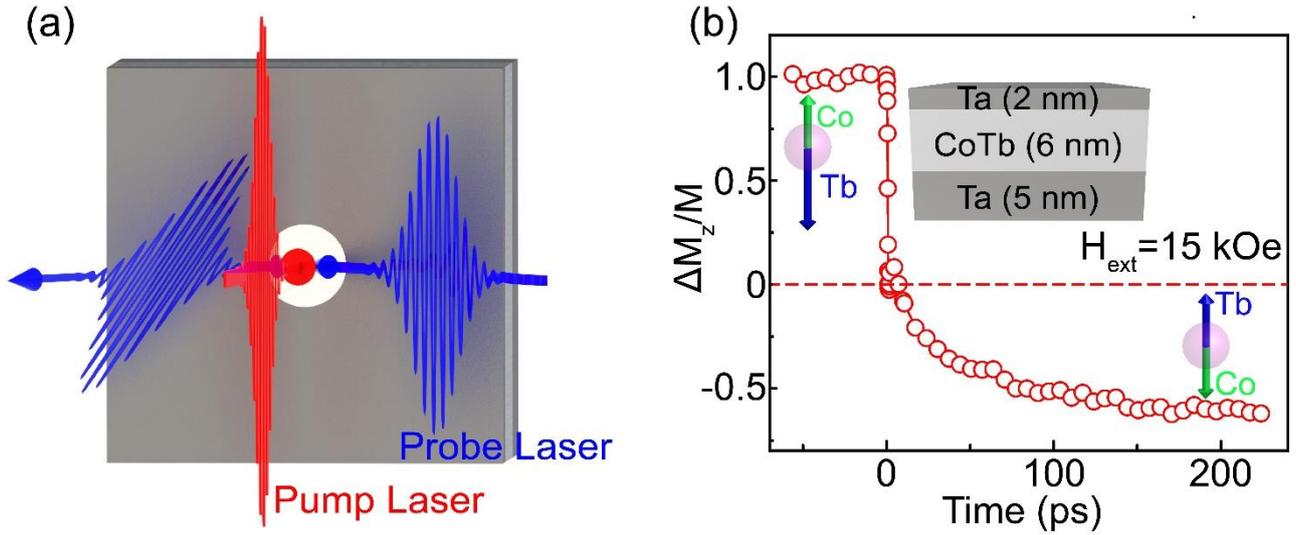


Fig. 1 (a) TRMOKE method is used to measure the ultrafast process of CoTb. (b) The ultrafast process of  $\text{Co}_{0.76}\text{Tb}_{0.24}$  alloy with an external magnetic field  $H_{\text{ext}}$  of 15 kOe.

To further examine this process,  $\text{Co}_{1-x}\text{Tb}_x$  alloys with different compositions were prepared. Fig. 2(a) shows the hysteresis loops of  $\text{Co}_{1-x}\text{Tb}_x$  with  $0.12 \leq x \leq 0.3$  measured by MOKE at room temperature. It has a laser with a wavelength of 400 nm, which can probe the magnetic moment of the Co sublattice. The coercivity field increases rapidly as the compensation composition  $x=0.2$  is approached. We observe a reversal of polarity of hysteresis loops at the compensation composition  $x \sim 0.2$ . The samples are Co-dominant for  $x < 0.2$  and Tb-dominant for  $x > 0.2$ . The ultrafast magnetization dynamics of the CoTb alloys near the compensation composition at room temperature are summarized in Figs. 2(b) and 2(c), which were all measured at an external magnetic field of 15 kOe. For the Co-dominant samples ( $x=0.14, 0.16, 0.18$ ), only ultrafast demagnetization can be observed. The Co sublattice relaxes back to its initial magnetization state after complete demagnetization, as shown in Fig. 2(b). For Tb-dominant samples ( $x=0.24, 0.26, 0.28, 0.30$ ), the Co sublattice reverses to the opposite magnetization state after full demagnetization, as shown in Fig. 2(c). For the alloy closest to the magnetization compensation composition ( $x=0.24$ ), the Co sublattice reverses 59% in the opposite magnetization state, while for the alloy farthest from the compensation composition ( $x=0.30$ ), the Co sublattice

reverses only 10% in the opposite magnetization state. The degree of switching decreases gradually with increasing the Tb concentration. The sample of  $x=0.22$  is Tb-dominant at room temperature. We observe ultrafast demagnetization, which appears irregular at first glance (Fig. 2(b)). We measured the hysteresis loop at a negative delay, shown as the inset in Fig. 2(b). The polarity of the loop under negative delay reverses compared with its loop measured in Fig. 2(a), and the coercivity field decreases. It indicates that the sample probed under negative delay has changed to Co-dominant. As the  $T_M$  of the sample ( $x=0.22$ ) is very close to room temperature, the residual heating from pump pulses is sufficient to change this sample from Tb-rich into Co-rich, which is supported by the reversal of loop polarity. Those results indicate the decisive role of  $T_M$ .

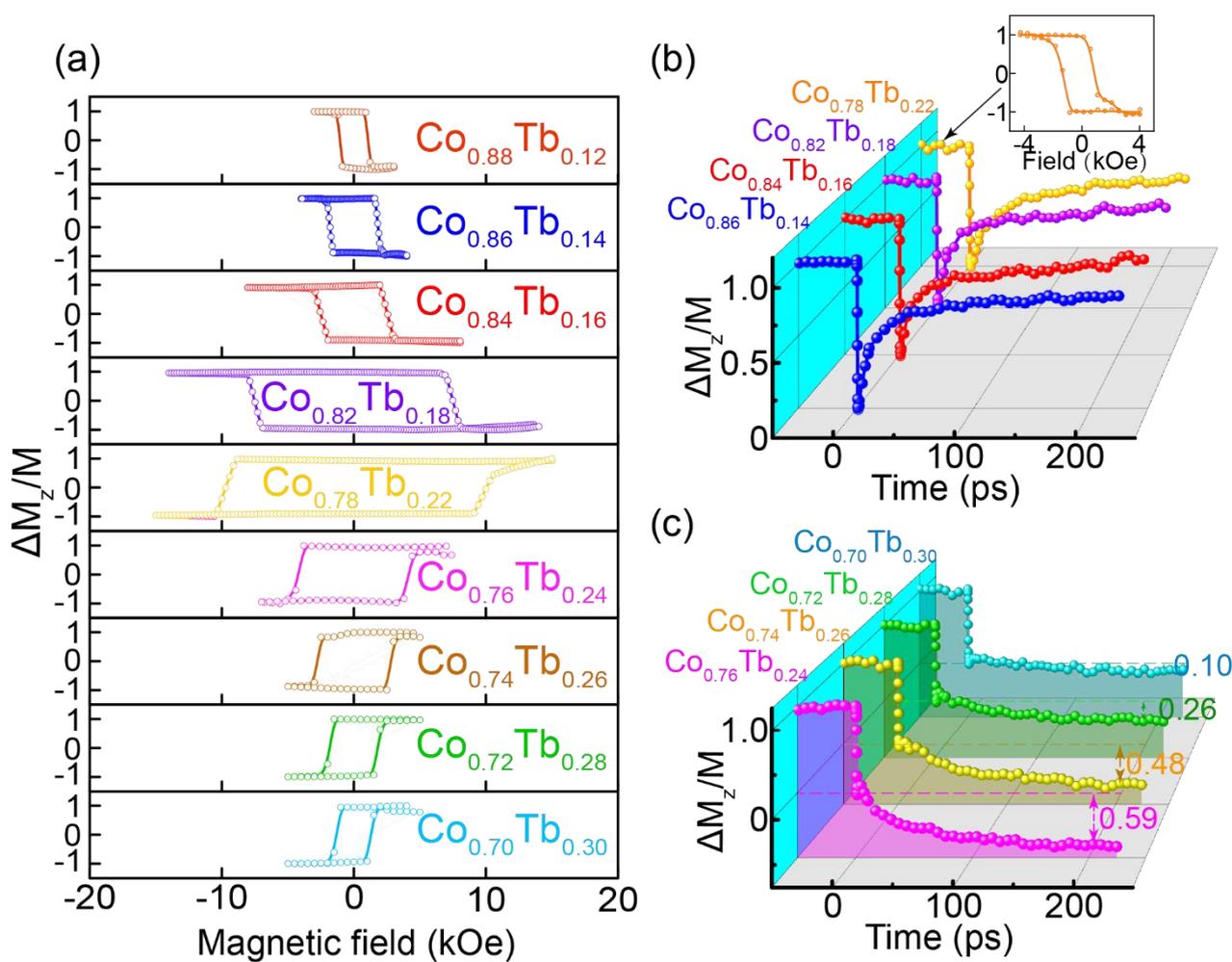


Fig.2 (a) Hysteresis loops of  $\text{Co}_{1-x}\text{Tb}_x$  thin films from MOKE with  $x$  varies from 0.12 to 0.3. Ultrafast magnetization dynamics of (b)  $\text{Co}_{1-x}\text{Tb}_x$  with  $x = 0.14, 0.16, 0.18, 0.22$ , and (c)  $\text{Co}_{1-x}\text{Tb}_x$  with  $x = 0.24, 0.26, 0.28, 0.30$ , which were all measured at an external magnetic field of 15 kOe.

The ultrafast magnetization dynamics of  $\text{Co}_{0.76}\text{Tb}_{0.24}$  and  $\text{Co}_{0.72}\text{Tb}_{0.28}$  at different pumping fluence were also studied, as shown in Figs. 3(a) and 3(b). An external magnetic field of 15 kOe was applied to them. At high pump fluence ( $7.9 \text{ mJ/cm}^2$ ), the Co sublattices of the two demagnetized rapidly to zero and then relaxed to

opposite magnetization state. At low pump fluence, the ultrafast magnetic dynamic processes of the two are different. For  $\text{Co}_{0.72}\text{Tb}_{0.28}$ , the Co sublattice relaxes towards the initial magnetization state after incomplete demagnetization at a lower pump fluence ( $3.9 \text{ mJ/cm}^2$ ), as shown in Fig. 3(b). While for  $\text{Co}_{0.76}\text{Tb}_{0.24}$ , the Co sublattice relaxes towards the opposite state after incomplete demagnetization at the same pump fluence ( $3.9 \text{ mJ/cm}^2$ ). This indicates that CoTb can initiate relaxation towards the opposite magnetization state without full demagnetization, which is different from GdFeCo alloys<sup>7</sup>. The magnetic dynamics process within 10 ps after ultrafast demagnetization was scanned in detail, as shown in the inset in Fig. 3(a). It is found that after ultrafast demagnetization, the magnetization recovered partially and then reversed to the opposite state. This switching process is slower than the GdFeCo, requiring at least 7 ps.

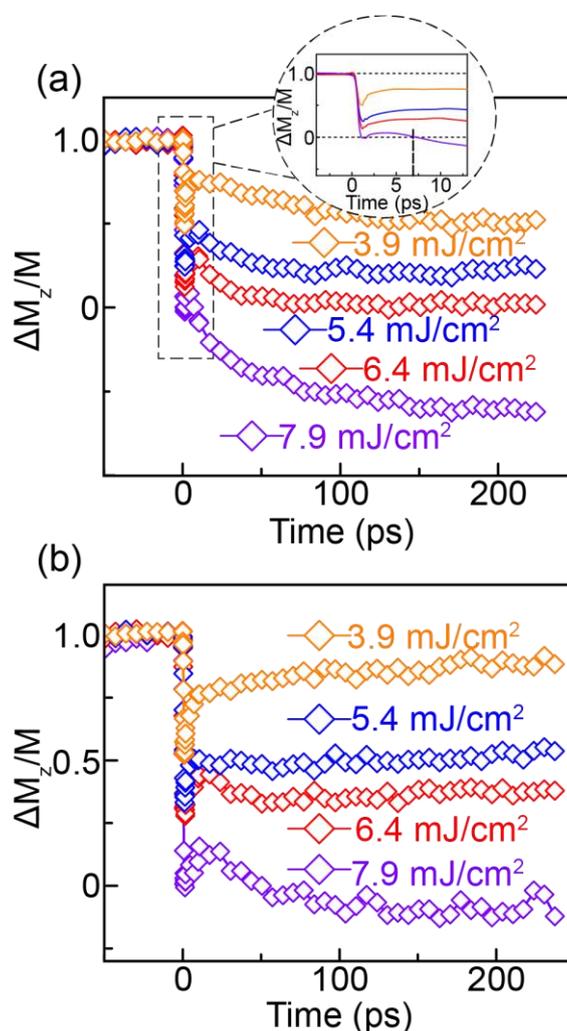


Fig. 3 Effect of fluence on switching process. (a) Ultrafast magnetization dynamics of  $\text{Co}_{0.76}\text{Tb}_{0.24}$  under different pumping fluences. (b) Ultrafast magnetization dynamics of  $\text{Co}_{0.72}\text{Tb}_{0.28}$  under different pumping fluences.

Finally, we studied the effect of the magnetic field on  $\text{Co}_{0.76}\text{Tb}_{0.24}$  at different pumping fluences. At lower

pumping fluence ( $3.9 \text{ mJ/cm}^2$ ), the relaxation process was significantly enhanced with the increase of magnetic field (Fig. 4(a)). At high fluence ( $7.9 \text{ mJ/cm}^2$ ), the relaxation process does not vary with the magnetic field (Fig. 4(b)). For  $\text{Co}_{0.72}\text{Tb}_{0.28}$ , the magnetic field has no effect on the relaxation process for both low ( $3.9 \text{ mJ/cm}^2$ ) and high fluences ( $7.9 \text{ mJ/cm}^2$ ), as seen respectively in Fig. 4(c) and Fig. 4(d).

To confirm the role of the magnetic field in the switching process, we performed the single pulse experiment on a  $\text{Co}_{0.76}\text{Tb}_{0.24}$  sample with a MOKE microscope, as shown in Fig. 4(e). The femtosecond laser repetition rate was reduced from 1 kHz to 1 Hz using a pulse picker, and the beam was then focused on the sample surface using an objective. A Light Emitting Diode (LED) with a wavelength of 623 nm is used as a light source for imaging. A Polarizer is used to obtain linearly polarized LED light. The LED light is focused on the surface of the sample by the objective. The reflected light passes through the analyzer and enters the Charge Coupled Device (CCD) camera by the beam splitting mirror to obtain the MOKE image. The magnetic field is applied with a permanent magnet during the measurement. The MOKE imaging is shown in the inset in Fig. 4(e). The first, second, and third rows correspond to the results after single pulse excitation with a field applied along +z, -z, and no field. We observe that magnetization direction is reversed upon a single pulse in the presence of an opposite field. Furthermore, no applied field results in a demagnetized state. This clearly demonstrates that the magnetic field is necessary for the switching.

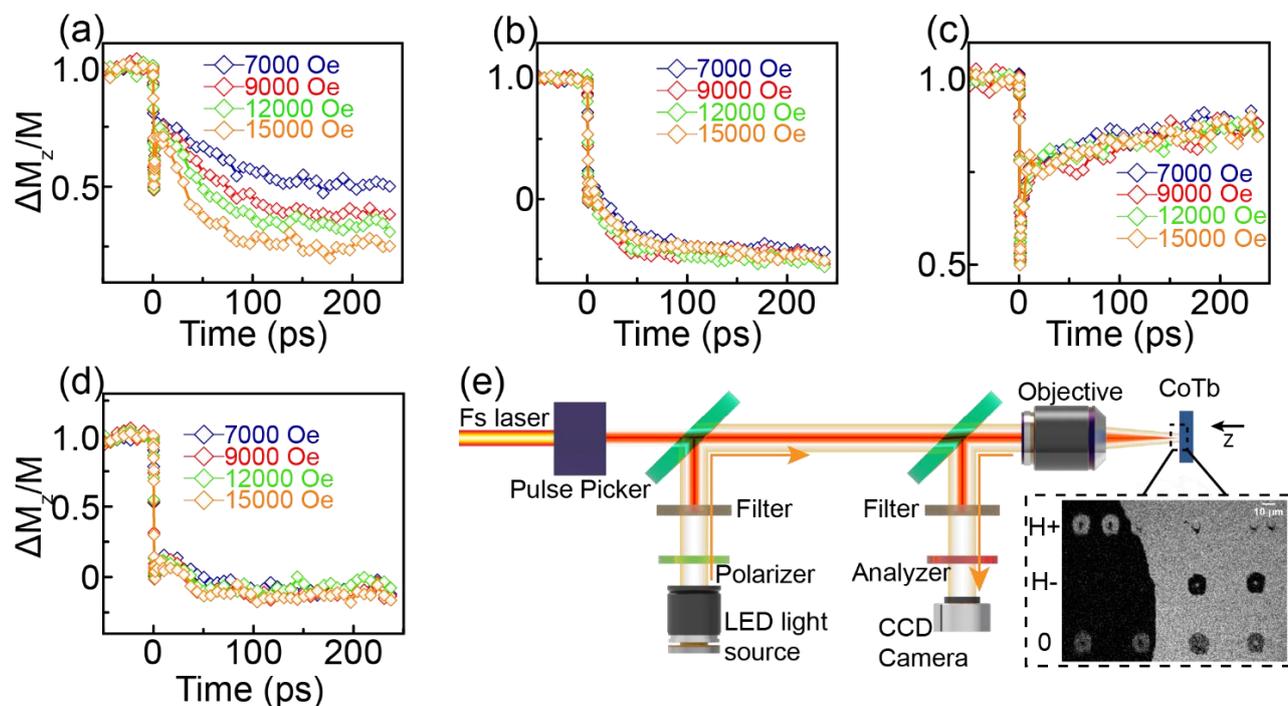


Fig. 4 Effect of magnetic field on switching process. Ultrafast process of  $\text{Co}_{0.76}\text{Tb}_{0.24}$  with different magnetic fields under (a)  $3.9 \text{ mJ/cm}^2$

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and (b) 7.9 mJ/cm<sup>2</sup>. (c) Ultrafast process of Co<sub>0.72</sub>Tb<sub>0.28</sub> with different magnetic field under (c) 3.9 mJ/cm<sup>2</sup> and (d) 7.9 mJ/cm<sup>2</sup> pump fluence. (e) MOKE imaging after single pulses with +10 Oe, -10 Oe, and no magnetic field with a fluence of 5.2 mJ/cm<sup>2</sup>.

The distinct magnetization dynamics for Tb- and Co-rich samples suggest the critical role of the compensation temperature. The compensation temperature should be reached at 3.9 mJ/cm<sup>2</sup> for only x=0.24. To estimate the compensation temperature, we simulated the temperature dependency of magnetization in our Co<sub>1-x</sub>Tb<sub>x</sub> alloy<sup>27</sup> with the parameters extracted from Moreno *et al.*<sup>28</sup> as explained in S1. Different compositions lead to different temperatures of magnetization compensation  $I$ , angular compensation ( $T_A$ ), and Curie temperatures ( $T_C$ ), as seen in Fig. 5(a). As expected, since the Tb-Tb coupling is lower than the Co-Co coupling, the compensation (resp. Curie) temperature increases (resp. decreases) with the Tb concentration. Since there is a 100 K difference between the compensation temperatures for x=0.24 and x=0.28, both can have a different field sensitivity at low laser power. Furthermore, there is no angular composition for x =0.28 since it reaches its  $T_C$  beforehand. It is not necessary to reach the angular compensation for the ultrafast switching.

To gain insight into the time on the timescale of temperature evolution in our CoTb samples, we used the two-temperature model (2TM) to describe their ultrafast behavior<sup>1</sup> with parameters detailed in S2. The obtained electrons and lattice temperatures ( $T_e$  and  $T_l$ ) are plotted in Fig. 5(b). After the initial rise of the  $T_e$ , it takes 4 ps for the electrons and the lattice to reach thermal equilibrium. We also indicated on the same graph the compensation temperature for x=0.24. For the simulated power used, the magnetic compensation is reached for x=0.24 but not for x=0.28, reproducing the low fluence regime with composition-dependent field sensitivity. The electron and lattice are in equilibrium at 4 ps.

The reversal occurs in several tens of picoseconds. The electron and lattice are in thermal equilibrium during the reversal. Such an ultrafast switching can occur either by domain wall propagation or coherent precession. Yet the domain wall velocity in the flow regime strongly depends on the magnetic field strength, while the reversal observed in Fig. 4(b) seems independent of the magnetic field strength. Therefore, we employed coherent precession to explain the possible switching dynamics, and performed macrospin LLG simulations under an applied field for x=0.24 with one global magnetic moment for Tb and one for Co with the expression in S3. The temperature is fixed for each simulation since we aim to reproduce the magnetization evolution once both  $T_e$  and  $T_l$  are at equilibrium. Both Co and Tb are considered aligned along the process so that no exchange term is included in the effective field used for the simulation. The expression used for the temperature dependence of the anisotropy can be found in literature<sup>7</sup>. We considered both damping parameters

equal to 0.03<sup>29</sup>. We focused our simulations around the  $T_A$  (585 K for  $x=0.24$ ) and plotted the switching time as a function of magnetic field at different temperatures in Fig. 5(c).

We observe that the switching time decreases at the  $T_A$  (585 K for  $x=0.24$ ). Furthermore, the switching time is in the picosecond range, in consistence with our TRMOKE measurements. We notice that a picosecond switching can still occur when the temperature is slightly below the angular compensation point. Most importantly, the switching time is almost constant once the magnetic field is high enough, as in Fig. 4. Thus, a coherent precession of the magnetization could explain the ultrafast magnetization switching of CoTb.

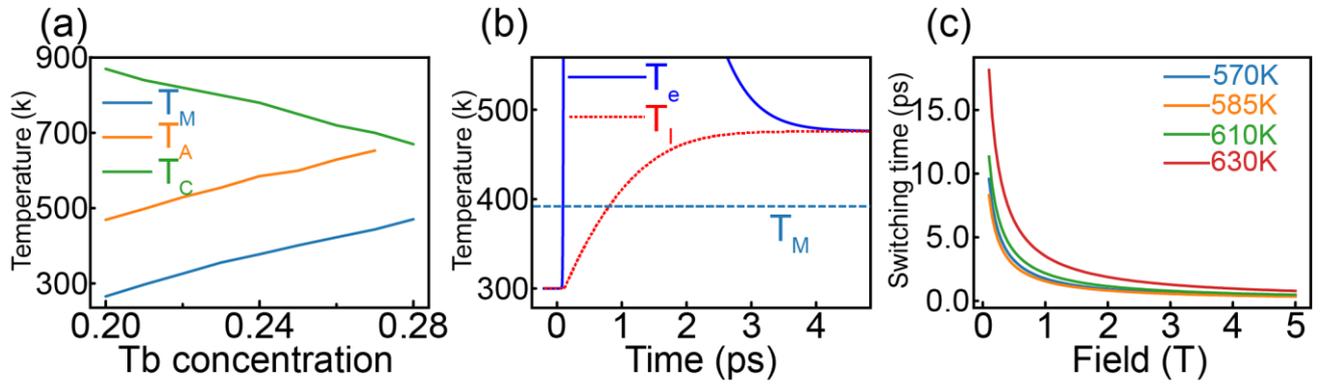


Fig. 5 Simulations of (a) the evolution of the Curie and compensation temperature with the Tb concentration, (b) electronic and lattice temperature evolution in the 2T model, (c) switching time as a function of the magnetic field for different temperatures.

In conclusion, we experimentally investigated the ultrafast magnetic switching process of  $\text{Co}_{1-x}\text{Tb}_x$  ferrimagnetic samples. We found that the magnetization dynamics are composition-sensitive, highlighting the role of compensation temperature for the picosecond reversal. Moreover, the magnetic field dependence suggests the switching is driven by a coherent precession of magnetization in the presence of an external field. Our results establish the single pulse switching of CoTb, which extends the potential materials for ultrafast magnetization control.

See supplementary material for more descriptions of simulation results.

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