Conditions for thermally induced all-optical switching in ferrimagnetic alloys: Modeling of TbCo

MORENO, Roberto, OSTLER, Thomas <http://orcid.org/0000-0002-1328-1839>, CHANTRELL, Roy and CHUBYKALO-FESENKO, Oksana

Conditions for thermally induced all-optical switching in ferrimagnetic alloys: Modeling of TbCo

R. Moreno,[1] T. A. Ostler,[2,3] R. W. Chantrell,[4] and O. Chubykalo-Fesenko[1]
[1]*Instituto de Ciencia de Materiales de Madrid, CSIC, Cantoblanco, 28049 Madrid, Spain*
[2]*Department of Physics, Université de Liège, B-4000 Liège, Belgium*
[3]*College of Engineering, Mathematics and Physical Sciences, Harrison Building, Streatham Campus, University of Exeter, North Park Road, Exeter, EX4 4QF, United Kingdom*
[4]*Department of Physics, University of York, Heslington, York YO10 5DD, United Kingdom*
(Received 26 January 2017; revised manuscript received 7 April 2017; published 6 July 2017)

We present atomistic spin dynamics modeling of thermally induced magnetization switching (TIMS) of disordered ferrimagnetic TbCo alloys varying the Tb concentration, laser pulse fluence, and its duration. Our results indicate that deterministic TIMS occurs in a wide range of Tb concentrations and at large laser fluences with a pulse duration of 50 fs. We furthermore demonstrate that the occurrence of the transient ferromagneticlike state is necessary, but after first reversal, the system may switch back. The presence of a magnetization compensation point or going through it is shown not to be required. With the increase of the laser pulse duration TIMS becomes stochastic so that for a 1 ps laser pulse width and beyond the deterministic heat-assisted AOS does not exist.

DOI: 10.1103/PhysRevB.96.014409

I. INTRODUCTION

In the last two decades the development and availability of means of exciting magnetic materials on the subpicosecond time scale, such as femtosecond laser pulses [1–3], THz [4,5], or mid-infrared sources, has given rise to a growing area of science broadly known as ultrafast spin dynamics. The interest in this field is on the one hand to understand the nonequilibrium processes and on the other to use this knowledge to develop next generation technologies that operate at much higher data rates than present day technology.

In 2007 Stanciu *et al.* reported that in GdFeCo disordered alloys it was possible, not only to change, but also to reverse magnetization using circularly polarized femtosecond laser pulses [6]. This observation lead to a debate as to the origin of the ultrafast switching [7,8]. The inverse Faraday effect (IFE) was originally proposed as the key driving mechanism for the reversal [6,9,10]. A further complication to the debate on all-optical switching (AOS) was presented in 2012 by Ostler *et al.* where single 50 fs laser pulses were shown to drive deterministic magnetization reversal for both left, right, and linearly polarized light [11]. This has been verified experimentally [11,12] in GdFeCo leading to the conclusion that there are indeed (at least) two mechanisms for AOS; one that is helicity dependent; and another that is purely thermally driven (dubbed thermally induced magnetization switching or TIMS) which is helicity independent. Typically the first mechanism requires multiple pulses while the second mechanism is feasible with only a single, femtosecond laser pulse.

A large amount of work in the area of AOS has been carried out in the prototypical disordered ferrimagnetic alloy GdFeCo of various compositions, using laser pulses in the regime from tens of femtoseconds, see, e.g., [6,11–14], and up to picoseconds [15,16]. However, there have also been a number of demonstrations of AOS in other ferrimagnets [17,18], ferromagnets [19], and multilayer combinations [20–22]. In some material systems a *single* pulse mechanism has been demonstrated [11], while in others multiple pulses have been shown to be required [7]. The studies have found that the helicity-dependent AOS most frequently requires a large number of repeated pulses, suggesting that the IFE effect is likely to be relatively small [7]. Alternatively, the magnetic circular dychroism (MCD) has been suggested as the microscopic mechanism for the helicity-dependent switching [12]. Recent calculations [23] have shown that the magnitudes of the IFE and MCD effects may be similar, both being the source for asymmetric spin-flip probability during the laser heating and thus we do not distinguish between them. Furthermore, the cumulative heating effect is likely to contribute to the reversal process where the system is driven close to the phase transition temperature. A further complication is that at the threshold for switching, the ultrafast TIMS in GdFeCo becomes helicity dependent even with one laser pulse showing that the two mechanisms coexist [12]. Additionally, on the larger time scale the asymmetry in spin-flip probabilities leads to nonsymmetric domain expansion, the domain sizes being proposed as the criterion for AOS [7,24].

In this article we center on the pure heat-assisted AOS, i.e., TIMS, previously confirmed in GdFeCo both experimentally [6,11–14] and theoretically [11,14,25,26]. TIMS is a deterministic phenomenon which does not require any asymmetry of the spin-flip probability coming from IFE, MCD, or external field. In Refs. [11,14] it was empirically demonstrated (see also discussion in Ref. [27]) that a number of requirements for TIMS must be satisfied to observe the phenomena. These include (i) two or more magnetic species with antiferromagnetic exchange, (ii) different demagnetization times, (iii) the presence and traversal of the magnetic compensation point T_M, and (iv) ultrashort laser pulses. As well as these requirements, it was also shown in Ref. [14] that the switching in GdFeCo proceeded through a temporary alignment of the Fe and Gd sublattices, though it is not clear if the existence of this *transient* state is a requirement or merely a side effect of the switching. In the single subpicosecond pulse mechanism the first two requirements have so far been unambiguously demonstrated. However, also rather long laser pulses have been used to drive magnetization reversal, up to 10 ps [18], raising fundamental questions about the intrinsic or limiting time scales of the underlying processes.

The common feature of AOS in ferrimagnetic materials is that it is observed experimentally in disordered materials

for a certain range of rare-earth concentration in the sample for which the magnetization compensation temperatures T_M exists. There has been a lot of discussion about the role of T_M (or close to it the angular momentum compensation point T_A) in AOS. One of the first ideas was that going through these points is crucial for AOS since at T_A the effective macroscopic damping diverges [6]. Recently, the modeling and experiments [11] have indicated that the occurrence of these points is not necessary. A more recent article [25] argues that the most energy-efficient switching occurs at temperatures close to this temperature and in general in the conditions of low magnetization [25,28]. This indicates that the role of the compensation point is still an open question.

While GdFeCo has been the subject of many theoretical studies, TbCo has been so far mostly investigated experimentally [7,17,18,20], motivating the need for a theoretical investigation of switching. Apart from experimental reports [29,30] on AOS, TbFe has also been modeled atomistically, though mostly from the point of view of static magnetization [31]. At the same time, both TbFe and TbCo present larger anisotropy than GdFe, and thus potentially are more relevant for applications. Recently nanoscale magnetic recording with AOS technology using near-field optics has been reported on TbFeCo thin films [32]. Most of the experimental studies on TbCo have been subject to switching through the use of circularly polarized laser pulses [7,17,20] that confirmed the occurrence of AOS in TbCo, but mainly the inverse-Faraday effect has been speculated as the mechanism. In a recent experiment on TbCo [7,18], with a single laser pulse, TIMS has not been confirmed, and only thermal demagnetization with subsequent formation of domains has been reported. Due to the fact that different experiments are performed in different conditions, i.e., different pulse lengths, fluences, static vs time-resolved probing, single-shot vs multiple shot laser pulses, and others, they cannot be considered as conclusive at this point and the possibility of TIMS in TbCo is an open question. Thus, addressing this problem theoretically and finding the region of parameters for which TIMS exists, if it does, is important to guide future experiments.

In order to investigate and clarify some of the aforementioned aspects of TIMS, such as the requirement of the presence (and traversal) of T_{M}, the transient ferromagneticlike state, and the importance of the laser pulse duration, a computational study of the magnetization dynamics for disordered TbCo alloys excited with ultrashort laser pulses is presented in this paper. TbCo has been selected here (many ferrimagnetic compounds with rare-earth and transition-metal elements could exhibit TIMS) for several key reasons. First, it has been demonstrated experimentally that AOS can be produced with different laser pulse durations [7,17,18]. Second, it is an interesting material for ultrahigh density data storage, and finally, the conditions for its switching under TIMS has not been investigated theoretically so far.

Specifically, here we investigate the influence of the Tb concentration x in the sample $\mathrm{Tb}_x\mathrm{Co}_{1-x}$ with different (or no) compensation temperatures, heated by laser pulses with different durations (50 fs, 400 fs, 1 ps, 10 ps) and fluences, and seek the occurrence of subpicosecond magnetization reversal without any other external stimulus (such as a magnetic field or spin polarized current). For this purpose, atomistic spin simulations of TbCo have been performed.

II. MODEL

Our model is based on the extended Heisenberg Hamiltonian assuming localized magnetic moments on both Tb and Co sites. The Hamiltonian reads

$$\mathcal{H} = -\frac{1}{2}\sum_{i\neq j} J_{ij}\mathbf{S}_i \cdot \mathbf{S}_j - \sum_i k_{\mathrm{u}} S_z^2, \tag{1}$$

where $\mathbf{S}_i$ are classical spins vectors with unit length describing the magnetic moment directions on site i. J_{ij} is the interatomic exchange interaction, and k_{u} is the local uniaxial anisotropy constant per atom. Previously this model has been shown to correctly describe ultrafast magnetization dynamics in GdFeCo [11]. The dynamics of the many-body spin system are calculated by integration of a stochastic Landau-Lifshitz-Gilbert equation on the atomic level:

$$\frac{d\mathbf{S}_i}{dt} = -\frac{\gamma_i}{\left(1+\lambda_i^2\right)\mu_i}\mathbf{S}_i \times [\mathbf{H}_i + \lambda_i \mathbf{S}_i \times \mathbf{H}_i], \tag{2}$$

where λ_i is the coupling to the thermal bath. The value of the magnetic moment is given by μ_i. $\mathbf{H}_i = -\partial\mathcal{H}_i/\partial\mathbf{S}_i + \boldsymbol{\zeta}_i$ is the effective field at site i and includes stochastic thermal fluctuations $\boldsymbol{\zeta}_i$. The thermostat is coupled to the thermal bath represented by the electronic temperature of the system (see details below).

To model the properties of TbCo we have used the following material parameters: damping constant $\lambda = 0.05$ for both materials, and magnetic moments $\mu_{\mathrm{Co}} = 1.61\mu_B$ [33] and $\mu_{\mathrm{Tb}} = 9.34\mu_{\mathrm{B}}$ [34]. The selected magnetic moment values for our simulations are the corresponding bulk values for Co and Tb. However, it is known that these values might change from pure metals to alloys, see, e.g., Ref. [35]. Nevertheless, the variation should be small, i.e., see Ref. [33] for cobalt, thus, we do not expect that this difference influences our results significantly. The on-site uniaxial magnetic anisotropies used in this work are $k_{\mathrm{u}}(\mathrm{Co}) = 3.73 \times 10^{-23}$ J/atom and $k_{\mathrm{u}}(\mathrm{Tb}) = 2.16 \times 10^{-22}$ J/atom. A different anisotropy constant was used for Co and Tb in order to mimic the change of macrosopic anisotropy as a function of Tb concentration, reported in Ref. [17], although we stress that the anisotropy does not play a significant role in the ultrafast switching. These values have similar magnitudes as published in Ref. [17]. The value of damping was chosen to give the magnetization quenching magnitudes similar to that observed in Ref. [18], as discussed below. The simulated system size was $50 \times 50 \times 50$ fcc atomic cells where Co was randomly substituted by Tb with the desired concentration.

As was done for GdFeCo [36], an empirical fitting to experimental measurements of the Curie temperature T_{C} and the magnetization compensation point T_{M} was carried out using the experimental values from Ref. [17] as a function of Tb concentration x. As a first step, bulk exchange parameters for Co ($J^{\mathrm{bulk}}_{\mathrm{Co\text{-}Co}}$) and Tb ($J^{\mathrm{bulk}}_{\mathrm{Tb\text{-}Tb}}$) corresponding to each bulk T_{C} (1398 K [37] and 237 K [38], respectively) have been used. The antiferromagnetic coupling value ($J^{\mathrm{bulk}}_{\mathrm{Co\text{-}Tb}}$) could be used to fit the experimental dependence $T_{\mathrm{M}}(x)$. However, the Curie temperatures T_{C}, obtained as a function of this parameter, are much higher than those measured experimentally. Here we include an extra antiferromagnetic exchange interaction in the Co sublattice (equivalent to the decrease of the Co-Co

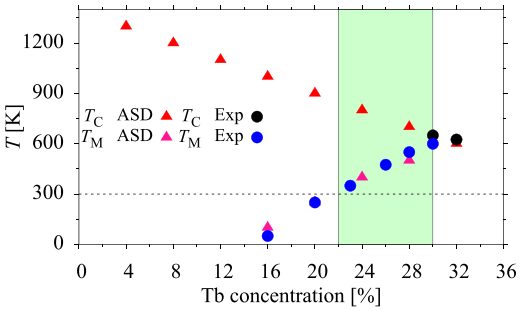

FIG. 1. Curie temperature T_C and the compensation temperature T_M as a function of Tb concentration x. Triangular point (▲/▲) symbols represent the results from atomistic spin dynamics (ASD) simulations with the parameters from Table I. Circles (●/●) represent the experimental values extracted from Ref. [17]. As the simulations begin at room temperature the white background is the concentration region where T_M is not reachable with the laser heating.

exchange) that appears in the presence of Tb ($J_{\text{Co-Tb-Co}}$) and is proportional to the Tb concentration (as in the mean field) and reduces the T_C value, as suggested in Refs. [18,39]. Therefore, the effective exchange parameter on the Co site reads ($x < 0.5$)

$$J^{\text{eff}}_{\text{Co-Co}} = J^{\text{bulk}}_{\text{Co-Co}} + J_{\text{Co-Tb-Co}} \frac{x}{1-x} \tag{3}$$

and the additional mean field acting on Co is proportional to the Tb concentration x. Using the two fitting parameters $J_{\text{Co-Tb}}$ and $J_{\text{Co-Tb-Co}}$, we can reproduce the experimental dependence of $T_M(x)$ and $T_C(x)$ for all x, see Fig. 1. The corresponding exchange interaction parameters used in the atomistic spin dynamics model are presented in Table I.

As discussed in Ref. [18], there exist three different regions for the position of T_M relative to room temperature $T_{\text{room}} = 300$ K. The first one corresponds to concentrations in the region from $x = 0.04$ to $x = 0.2$ where T_M is below T_{room} or does not exist due to the shortage of Tb. Thus the transient temperature of the electronic bath will not cross T_M upon laser heating. The second one ranges from $x = 0.2$ to $x = 0.3$ and T_M is above T_{room} so that the electronic temperature may cross T_M upon application of the laser pulse, depending on the fluence. The third region begins at the Tb composition $x = 0.3$ where the compensation temperature T_M does not exist due to an excess of Tb. Considering this distinction, the role of T_M in the occurrence of AOS will be determined in the following.

In order to model the laser heating of the sample and the electronic temperature dynamics, the two-temperature model (2TM) is used. The pump energy is described using an exponential profile in time that allows us to vary the pulse duration of the laser. The 2TM therefore reads

$$\begin{aligned} C_e \frac{dT_e(t)}{dt} &= -G[T_e(t) - T_{\text{ph}}(t)] + P(t), \\ C_p \frac{dT_p(t)}{dt} &= G[T_e(t) - T_{\text{ph}}(t)], \\ P(t) &= (I_0 F) e^{-(t/\tau_p)^2}. \end{aligned} \tag{4}$$

Here $T_e(t)$ is the electronic temperature, $T_{\text{ph}}(t)$ is the temperature assigned to the phonon bath, $C_e = \gamma T_e$ and C_p are the heat capacities for electrons and phonons, and G is a coupling parameter between these systems. In the simulations we have used the following parameters taken from Ref. [11] for GdFeCo: $\gamma = 7.00 \times 10^2\ \text{J m}^{-3}\ \text{K}^{-2}$, $C_p = 3.0 \times 10^6\ \text{J m}^{-3}\ \text{K}^{-1}$, and $G = 17 \times 10^{17}\ \text{W m}^{-3}\ \text{K}^{-1}$. $P(t)$ is assumed to be the pump laser energy received by the electronic system, I_0 describes the amount of laser energy absorbed by the sample, F is the laser fluence, and τ_p is the laser pulse duration. More specifically, the spin dynamics simulations use the value $(I_0 F)$ characterizing the input energy [units of $\text{J}/(\text{m}^3\ \text{s})$], while the experimental papers report the values in terms of F (units J/m^2). The estimation of I_0 is nontrivial and the results of the simulations are strongly dependent on P. I_0 can be estimated from theory or experimental reflectivity, and has the units of $\text{m}^{-1}\ \text{s}^{-1}$. In this paper and because the experimental data are not available, its value has been taken from literature for FePt ($I_0 = 3 \times 10^{19}\ \text{m}^{-1}\ \text{s}^{-1}$) [40] giving laser pulse fluences in agreement with the experimental data on TbCo. We note that the results, while dependent on I_0, will be simply rescaled should a different value be used. This shows that this value, although somewhat arbitrary, is close to the real one for ultrathin metallic films. In the present work the simulations always start at room temperature $T_{\text{room}} = 300$ K.

TABLE I. Exchange parameters for TbCo, extracted by systematic variation of the Co-Tb and Co-Tb-Co parameters and comparing the T_C and T_M values with experimental measurements as a function of the concentration of Tb, x in the $\text{Tb}_x\text{Co}_{1-x}$ alloy.

Exchange interaction type	Energy (J)
$J^{\text{bulk}}_{\text{Co-Co}}$	5.9×10^{-21}
$J^{\text{bulk}}_{\text{Tb-Tb}}$	8.2×10^{-22}
$J_{\text{Co-Tb}}$	-1.0×10^{-21}
$J_{\text{Co-Tb-Co}}$	-4.4×10^{-21}

III. RESULTS

Figure 2 presents an example of simulated magnetization dynamics, in this case for $\text{Tb}_{32}\text{Co}_{68}$ and laser pulse duration

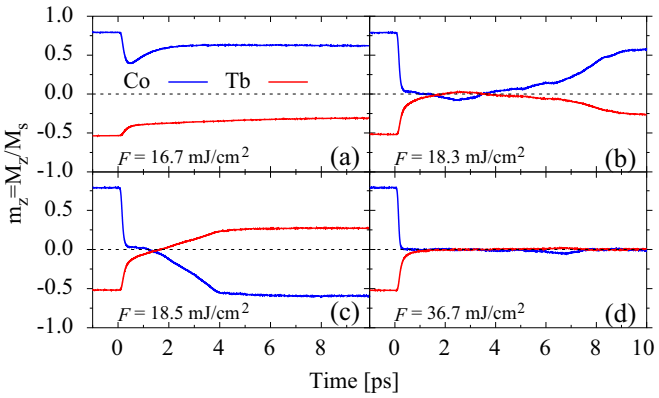

FIG. 2. Different magnetization dynamics curves corresponding to various pump fluences for $\text{Tb}_{32}\text{Co}_{68}$ and a pulse duration of 50 fs. The reduced magnetization $m_z = M_z/M_s$ is the magnetization of the individual sublattice divided by the value of that sublattice if all spins were ordered (the zero K case).

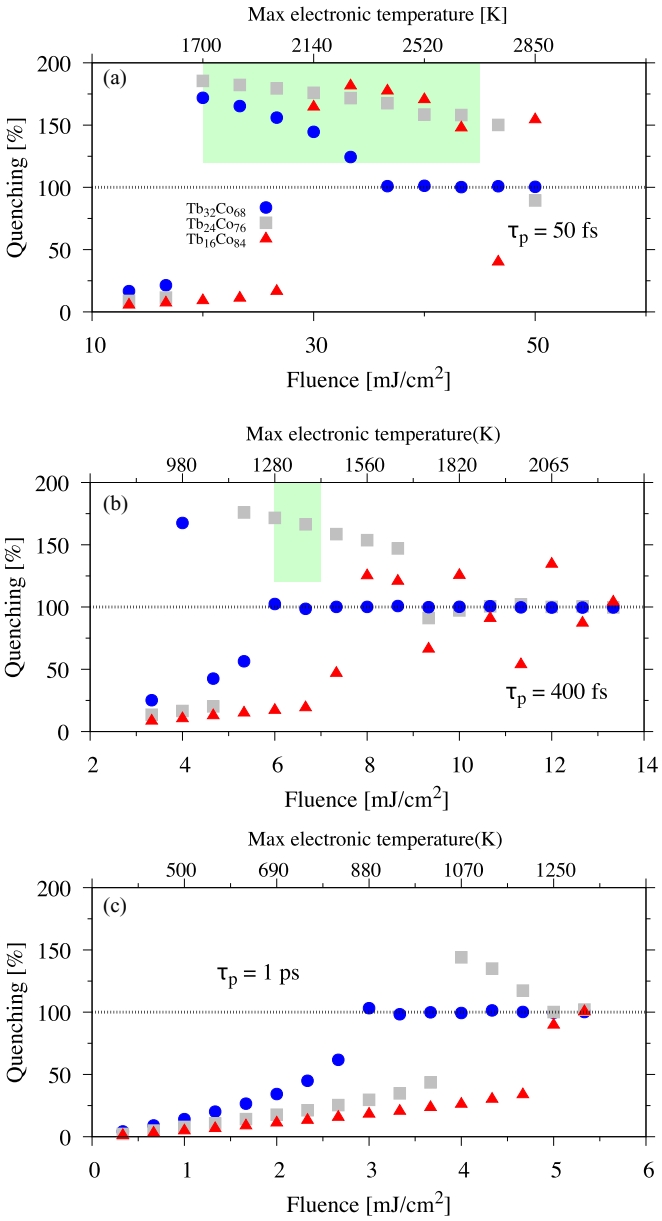

FIG. 3. Degree of magnetization quenching (z component) of the reduced magnetization (m_z) of Co 10 ps after the pulse for different compositions of TbCo, pump fluences, and laser pulse durations (a) 50 fs, (b) 400 fs, and (c) 1 ps. The green color indicates approximately the region with deterministic reversal.The numbers in the upper horizontal axis indicate maximum electronic temperature achieved in the integration of the $2T$ model.

$\tau_p = 50$ fs. Note that this composition does not have a magnetization compensation point. Our modeling qualitatively reproduces the experimental forms of the magnetization quenching/recovery and switching curves [18]. As expected, for small laser fluence [Fig. 2(a)] magnetization quenching and subsequent recovery takes place. For laser fluences above $F \sim 18.5\,\mathrm{mJ/cm^2}$ the magnetization of the sublattices switches, going through a small transient ferromagneticlike state [Fig. 2(c)], where the two sublattices temporarily align. We have investigated the fluence limit for which the switching starts to appear (see Fig. 3) finding that near the threshold value the magnetization can reach the transient ferromagnetic state and temporarily switch, however, a backward switching to the initial directions of the sublattices occurs, see Fig. 2(b). This indicates that the occurrence of the transient ferromagneticlike state is not a sufficient condition for switching. This form of the curve is similar to the experimentally measured one for TbCo [18] and TbFe [29]. At high laser power, the system simply demagnetizes as shown in Fig. 2(d).

In Fig. 3 we present quantitatively the degree of magnetization quenching for the z component of the Co sublattice magnetization after 10 ps, for different durations of the pulse (50 fs, 400 fs, 1 ps), and various compositions. In this figure, 100% quenching corresponds to demagnetization. Switching is represented as a magnetization quenching larger than 100%. The green shaded region represents deterministic switching (see below). A similar representation was used in Ref. [18]. The choice of compositions corresponds to $Tb_{16}Co_{84}$ (▲) with $T_M < T_{room}$, $Tb_{24}Co_{76}$ with $T_M > T_{room}$ (■), and $Tb_{32}Co_{68}$ (●) without a compensation point. The pump fluence in Fig. 3 has very different energy scales due to the fact that as the laser pulse duration is increased, the total energy absorbed in the system is also increased and thus less laser fluence is necessary to produce the same demagnetization. We can also see that as the Tb concentration increases, the degree of quenching is reduced. Simulations show the existence of TIMS for all temporal widths of laser pulse, though it can be seen from Fig. 3 that as the pulse becomes shorter, the range of compositions and laser fluences with TIMS becomes larger.

We should point out explicitly that for a laser pulse duration of 50 fs, all compositions shown in Fig. 3 switch. Since the composition $Tb_{32}Co_{68}$ does not have a magnetization compensation point, this demonstrates that the occurrence of the compensation temperature is not a necessary condition for TIMS. Our simulations show that the composition $Tb_{12}Co_{88}$, also without a compensation temperature, presents TIMS as well. Furthermore, for the 50 fs laser pulse duration, the range for TIMS also includes $Tb_{16}Co_{84}$, for which T_M is below T_{room}. Thus, going through this point is also not a necessary condition for the AOS. However, while we have demonstrated that T_M is not an essential criteria for switching, the results so far do not confirm that it is an irrelevant parameter in the occurrence of AOS. In fact, since we clearly observe that the composition $Tb_{16}Co_{84}$ required more laser energy to switch, the presence of T_M influences the switching process. Furthermore, the composition with a compensation temperature higher than room temperature (crossed by the electronic temperature under the action of the laser pulse), i.e., $Tb_{24}Co_{76}$, is the only one that undergoes switching for all laser pulse durations.

Finally, for $Tb_{32}Co_{68}$ and a laser pulse with a 400 fs duration [Fig. 3(b)], TIMS takes place in a small window of pump fluence, ending with a simple demagnetization at large fluences. There is also a random event of switching at small laser pulse fluency ($4\,\mathrm{mJ/cm^2}$). This indicates a possible stochasticity of the switching in this case which we will investigate in the following. To this end, in Fig. 4 we show the reversal probability as a function of laser pulse fluence, with the averaging of many realizations with different seeds for the random number generator that is used for the fluctuating stochastic field. The data are shown for various Tb concentrations and for 50 and 400 fs laser pulses. The figure

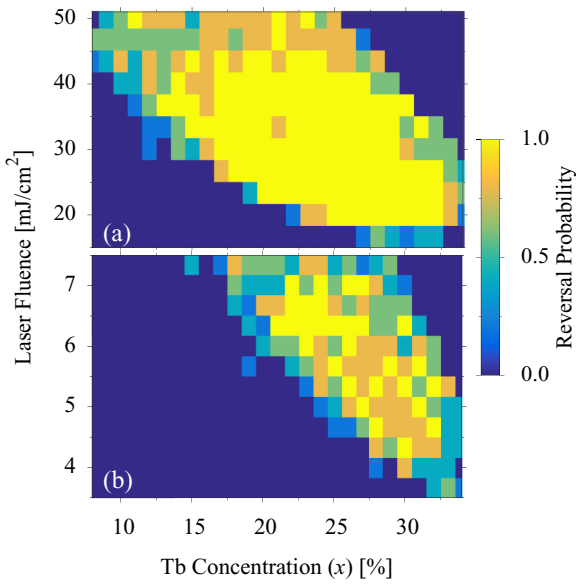

FIG. 4. Reversal probability for Tb_xCo_{1-x} alloys heated with different laser pump fluences. The color legend indicates the reversal probability with yellow and blue pixels reflecting the cases where there is magnetization switching or not, respectively. Upper a) and lower b) panels correspond to 50 fs and 400 fs laser pulse durations, respectively.

shows that for 50 fs laser pulses, deterministic TIMS in a wide range of Tb concentrations (x) is possible, with stochastic switching taking place around this region. The deterministic region is very small for the 400 fs laser pulse duration and does not exist for 1 ps laser pulses (not shown). This confirms that a fully deterministic TIMS only happens when the laser pulse duration is of the order of the characteristic time of the exchange interactions. For 50 fs laser pulses the deterministic region includes Tb concentrations above 30% for which T_M does not exist. The same is true for Tb concentrations $x < 0.2$ for which T_M is below T_{room} and thus it is not crossed during the heating process. Note that both the stochastic switching and complete demagnetization lead on larger time scale to the formation of magnetic domains, see previous modeling [41]. In the case of the stochastic switching, parts of the system switch and others do not, leading to the formation of domains. In the case of the complete demagnetization, the system recovers by means of creation of such domains. The domain sizes in the latter case are smaller than in the former case which can be larger than the laser spot as is discussed in Ref. [42].

We should note that for GdFeCo, pulses, longer than 1 ps, have been used to induce switching [15,16]. Though, in the work of Gorchon *et al.* [16] an electrical stimulus rather than an optical one was used and it remains an open and interesting question as to whether the same thermally induced stimulus is at work here. Using similar modeling as presented here, switching in GdFeCo via TIMS can be induced using laser pulses up to 1 ps [43], though the main reason for the difference with the present work is that in GdFeCo the damping constant is lower, which has the effect of *slowing down* the dynamics. The low damping constant is the result of the fact that the orbital angular momentum quantum number $L = 0$ for Gd f electrons which constitutes the main difference between Gd-based and Tb-based alloys.

IV. CONCLUSIONS

We have modeled ultrafast magnetization dynamics in Tb_xCo_{1-x} ferrimagnets varying the Tb concentration, laser pulse fluence, and laser pulse duration. The inclusion of additional antiferromagnetic exchange on Co sites, with the strength proportional to the Tb concentration (consistent with the assumption of previous works [18]), has allowed us to reproduce the experimental Curie and the magnetization compensation temperatures, the key factor for comparison with experimental measurements. We have modeled and analyzed the occurrence of TIMS in this system, shedding light on some previously suggested requirements for the switching mechanism.

By systematically varying the laser fluence and Tb concentration for different laser pulse durations we have found the region of parameters for which TbCo switches. Our results indicate that deterministic TIMS occurs for 50 fs laser pulses in a range of Tb concentrations and high laser pump fluences. At the boundaries for this switching TIMS is stochastic. Our results show that the region for TIMS decreases with the increase of laser pulse duration and the TIMS becomes a stochastic phenomenon. Thus, TIMS occurs for laser pulses with durations on the time scale of the exchange interactions. Indeed, using the uncertainly principle $\Delta t \Delta E \geqslant \hbar/2$, and a laser duration 50 fs, gives an energy of $\sim 10^{-21}$ J, which matches with the AFM exchange interaction. Therefore, we believe that the AOS found in Refs. [7,18] with 400 fs and 10 ps laser pulses most probably does not have a complete heating origin. In our view, in this case, similar to ferromagnetic FePt [19,23], the inverse Faraday or MCD effects produce an asymmetry in the nucleation of TIMS and stabilize the stochastic reversal producing a deterministic AOS with many laser pulses. The reason that TIMS has not been found so far in TbCo with a single fs laser pulse may reside in the fact that it requires high laser intensities. Indeed, comparing with the results of Ref. [7], our threshold values are at the border of the largest experimental ones.

Furthermore, we have observed that the occurrence of the transient ferromagneticlike state is a necessary but not a sufficient condition for the switching since with the laser power at the switching boundary, we present examples where this state occurs but the system switches back. We also confirm that the existence of the magnetization compensation point is not necessary for switching, nor is it necessary to go through it during the laser heating, since for both cases we have presented counterexamples. These results are in agreement with experiments on TbFe as shown in Ref. [29].

Finally, we note that the complete set of conditions for the TIMS is still elusive, most likely due to the fact that too many parameters simultaneously play a role in determining the magnetization dynamics, e.g., the exchange, the damping, and the magnetic moments [44]. We believe that in the present work we have made a step forward in this respect by establishing that (i) different demagnetizing rates of the materials and (ii) femtosecond laser pulse durations are necessary conditions. At the same time (i) the presence of the magnetization compensation point or heating through it is not a necessary condition for switching and (ii) the transient ferromagneticlike state is a necessary but not sufficient condition since reversal can be stochastic on the boundaries of the reversed region.

ACKNOWLEDGMENTS

This work was supported by the Spanish Ministry of Economy and Competitiveness under the Grants MAT2013-47078-C2-2-P, FIS2016-78591-C3-3-R, and by the European Community's Seventh Framework Programme (FP7/2007-2013) under grant agreement No. 281043, FEMTOSPIN. T. A. Ostler gratefully acknowledges the Marie Curie incoming BeIPD-COFUND fellowship program at the University of Liège.

[1] J.-Y. Bigot, M. Vomir, and E. Beaurepaire, Coherent ultrafast magnetism induced by femtosecond laser pulses, Nat. Phys. **5**, 515 (2009).

[2] E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot, Ultrafast Spin Dynamics in Ferromagnetic Nickel, Phys. Rev. Lett. **76**, 4250 (1996).

[3] A. Stupakiewicz, K. Szerenos, D. Afanasiev, A. Kirilyuk, and A. V. Kimel, Ultrafast nonthermal photo-magnetic recording in a transparent medium, Nature (London) advance on **542**, 71 (2017).

[4] T. Kampfrath, A. Sell, G. Klatt, A. Pashkin, S. Mährlein, T. Dekorsy, M. Wolf, M. Fiebig, A. Leitenstorfer, and R. Huber, Coherent terahertz control of antiferromagnetic spin waves, Nat. Photon. **5**, 31 (2011).

[5] T. Kubacka, J. A. Johnson, M. C. Hoffmann, C. Vicario, S. de Jong, P. Beaud, S. Grübel, S.-W. Huang, L. Huber, L. Patthey, Y.-D. Chuang, J. J. Turner, G. L. Dakovski, W.-S. Lee, M. P. Minitti, W. Schlotter, R. G. Moore, C. P. Hauri, S. M. Koohpayeh, V. Scagnoli, G. Ingold, S. L. Johnson, and U. Staub, Large-amplitude spin dynamics driven by a THz pulse in resonance with an electromagnon, Science (NY) **343**, 1333 (2014).

[6] C. D. Stanciu, F. Hansteen, A. V. Kimel, A. Kirilyuk, A. Tsukamoto, A. Itoh, and T. Rasing, All-Optical Magnetic Recording with Circularly Polarized Light, Phys. Rev. Lett. **99**, 047601 (2007).

[7] M. S. El Hadri, P. Pirro, C. H. Lambert, S. Petit-Watelot, Y. Quessab, M. Hehn, F. Montaigne, G. Malinowski, and S. Mangin, Two types of all-optical magnetization switching mechanisms using femtosecond laser pulses, Phys. Rev. B **94**, 064412 (2016).

[8] M. O. A. Ellis and R. W. Chantrell, Switching times of nanoscale FePt: Finite size effects on the linear reversal mechanism, Appl. Phys. Lett. **106**, 162407 (2015).

[9] A. V. Kimel, A. Kirilyuk, P. A. Usachev, R. V. Pisarev, A. M. Balbashov, and Th Rasing, Ultrafast non-thermal control of magnetization by instantaneous photomagnetic pulses, Nature (London) **435**, 655 (2005).

[10] S. Alebrand, A. Hassdenteufel, D. Steil, M. Cinchetti, and M. Aeschlimann, Interplay of heating and helicity in all-optical magnetization switching, Phys. Rev. B **85**, 092401 (2012).

[11] T. A. Ostler, J. Barker, R. F. L. Evans, R. W. Chantrell, U. Atxitia, O. Chubykalo-Fesenko, S. El Moussaoui, L. Le Guyader, E. Mengotti, L. J. Heyderman, F. Nolting, A. Tsukamoto, A. Itoh, D. Afanasiev, B. A. Ivanov, A. M. Kalashnikova, K. Vahaplar, J. Mentink, A. Kirilyuk, T. Rasing, and A. V. Kimel, Ultrafast heating as a sufficient stimulus for magnetization reversal in a ferrimagnet, Nat. Commun. **3**, 666 (2012).

[12] A. R. Khorsand, M. Savoini, A. Kirilyuk, A. V. Kimel, A. Tsukamoto, A. Itoh, and T. Rasing, Role of Magnetic Circular Dichroism in All-Optical Magnetic Recording, Phys. Rev. Lett. **108**, 127205 (2012).

[13] L. Le Guyader, M. Savoini, S. El Moussaoui, M. Buzzi, A. Tsukamoto, A. Itoh, A. Kirilyuk, T. Rasing, A. V. Kimel, and F. Nolting, Nanoscale sub-100 picosecond all-optical magnetization switching in GdFeCo microstructures, Nat. Commun. **6**, 5839 (2015).

[14] I. Radu, K. Vahaplar, C. Stamm, T. Kachel, N. Pontius, H. A. Dürr, T. A. Ostler, J. Barker, R. F. L. Evans, R. W. Chantrell, A. Tsukamoto, A. Itoh, A. Kirilyuk, T. Rasing, A. V. Kimel, and H. A. Dürr, Transient ferromagnetic-like state mediating ultrafast reversal of antiferromagnetically coupled spins, Nature (London) **472**, 205 (2011).

[15] K. Vahaplar, A. M. Kalashnikova, A. V. Kimel, S. Gerlach, D. Hinzke, U. Nowak, R. Chantrell, A. Tsukamoto, A. Itoh, A. Kirilyuk, and T. Rasing, All-optical magnetization reversal by circularly polarized laser pulses: Experiment and multiscale modeling, Phys. Rev. B **85**, 104402 (2012).

[16] J. Gorchon, R. B. Wilson, Y. Yang, A. Pattabi, J. Y. Chen, L. He, J. P. Wang, M. Li, and J. Bokor, Role of electron and phonon temperatures in the helicity-independent all-optical switching of GdFeCo, Phys. Rev. B **94**, 184406 (2016).

[17] S. Alebrand, M. Gottwald, M. Hehn, D. Steil, M. Cinchetti, D. Lacour, E. E. Fullerton, M. Aeschlimann, and S. Mangin, Light-induced magnetization reversal of high-anisotropy TbCo alloy films, Appl. Phys. Lett. **101**, 162408 (2012).

[18] S. Alebrand, U. Bierbrauer, M. Hehn, M. Gottwald, O. Schmitt, D. Steil, E. E. Fullerton, S. Mangin, M. Cinchetti, and M. Aeschlimann, Subpicosecond magnetization dynamics in TbCo alloys, Phys. Rev. B **89**, 144404 (2014).

[19] C.-H. Lambert, S. Mangin, B. S. D. C. S. Varaprasad, Y. K. Takahashi, M. Hehn, M. Cinchetti, G. Malinowski, K. Hono, Y. Fainman, M. Aeschlimann, and E. E. Fullerton, All-optical control of ferromagnetic thin films and nanostructures, Science **345**, 1337 (2014).

[20] S. Mangin, M. Gottwald, C.-H. Lambert, D. Steil, V. Uhlír, L. Pang, M. Hehn, S. Alebrand, M. Cinchetti, G. Malinowski, Y. Fainman, M. Aeschlimann, and E. E. Fullerton, Engineered materials for all-optical helicity-dependent magnetic switching, Nat. Mater. **13**, 286 (2014).

[21] C. Xu, T. A. Ostler, and R. W. Chantrell, Thermally induced magnetization switching in Gd/Fe multilayers, Phys. Rev. B **93**, 054302 (2016).

[22] R. F. L. Evans, T. A. Ostler, I. Radu, T. Rasing, and R. W. Chantrell, Ultrafast thermally induced magnetic switching in synthetic ferrimagnets, Appl. Phys. Lett. **104**, 082410 (2014).

[23] R. John, M. Berritta, D. Hinzke, C. Müller, T. Santos, H. Ulrichs, P. Nieves, J. Walowski, R. Mondal, O. Chubykalo-Fesenko, J. McCord, P. M. Oppeneer, U. Nowak, and M. Münzenberg, Magnetization switching of fept nanoparticle recording medium by femtosecond laser pulses, arXiv:1606.08723.

[24] R. Medapalli, D. Afanasiev, D. Kim, Y. Quessab, S. A. Monotoya, A. Kirilyuk, T. Rasing, A. V. Kimel, and E. E.

Fullerton, Mechanism of all-optical control of ferromagnetic multilayers with circularly polarized light, arXiv:1607.02505.

[25] J. Barker, U. Atxitia, T. A. Ostler, O. Hovorka, R. W. Chantrell, and O. Chubykalo-Fesenko, Two-magnon bound state causes ultrafast thermally induced magnetisation switching, Sci. Rep. **3**, 3262 (2013).

[26] O. J. Suarez, P. Nieves, D. Laroze, D. Altbir, and O. Chubykalo-Fesenko, Ultrafast relaxation rates and reversal time in disordered ferrimagnets, Phys. Rev. B **92**, 144425 (2015).

[27] A. V. Kimel, All-optical switching: Three rules of design, Nat. Mater. **13**, 225 (2014).

[28] A. Hassdenteufel, J. Schmidt, C. Schubert, B. Hebler, M. Helm, M. Albrecht, and R. Bratschitsch, Low-remanence criterion for helicity-dependent all-optical magnetic switching in ferrimagnets, Phys. Rev. B **91**, 104431 (2015).

[29] A. R. Khorsand, M. Savoini, A. Kirilyuk, A. V. Kimel, A. Tsukamoto, A. Itoh, and T. Rasing, Element-Specific Probing of Ultrafast Spin Dynamics in Multisublattice Magnets with Visible Light, Phys. Rev. Lett. **110**, 107205 (2013).

[30] A. Hassdenteufel, B. Hebler, C. Schubert, A. Liebig, M. Teich, M. Helm, M. Aeschlimann, M. Albrecht, and R. Bratschitsch, Thermally assisted all-optical helicity dependent magnetic switching in amorphous Fe_{100-x} Tb_x alloy films, Adv. Mater. **25**, 3122 (2013).

[31] J. Xiankai, Z. Zongzhi, and L. Yaowen, Modeling of temperature dependence of magnetization in TbFe films - an atomistic spin simulation study, SPIN **06**, 1650003 (2016).

[32] T. Liu, T. Wang, A. H. Reid, M. Savoini, X. Wu, B. Koene, P. Granitzka, C. E. Graves, D. J. Higley, Z. Chen, G. Razinskas, M. Hantschmann, A. Scherz, J. Stöhr, A. Tsukamoto, B. Hecht, A. V. Kimel, A. Kirilyuk, T. Rasing, and H. A. Dürr, Nanoscale confinement of all-optical magnetic switching in TbFeCo - competition with nanoscale heterogeneity, Nano Lett. **15**, 6862 (2015).

[33] S. Chadov, J. Minár, M. I. Katsnelson, H. Ebert, D. Ködderitzsch, and A. I. Lichtenstein, Orbital magnetism in transition metal systems: The role of local correlation effects, Europhys. Lett. **82**, 37001 (2008).

[34] K. M. Döbrich, G. Bihlmayer, K. Starke, J. E. Prieto, K. Rossnagel, H. Koh, E. Rotenberg, S. Blügel, and G. Kaindl, Electronic band structure and fermi surface of ferromagnetic Tb: Experiment and theory, Phys. Rev. B **76**, 035123 (2007).

[35] P. Hansen, C. Clausen, G. Much, M. Rosenkranz, and K. Witter, Magnetic and magneto optical properties of rare earth transition metal alloys containing Gd, Tb, Fe, Co, J. Appl. Phys. **66**, 756 (1989).

[36] T. A. Ostler, R. F. L. Evans, R. W. Chantrell, U. Atxitia, O. Chubykalo-Fesenko, I. Radu, R. Abrudan, F. Radu, A. Tsukamoto, A. Itoh, A. Kirilyuk, T. Rasing, and A. V. Kimel, Crystallographically amorphous ferrimagnetic alloys: Comparing a localized atomistic spin model with experiments, Phys. Rev. B **84**, 024407 (2011).

[37] I. Turek, J. Kudrnovsky, V. Drchal, P. Bruno, and S. Blügel, Ab initio theory of exchange interactions in itinerant magnets, Phys. Status Solidi (b) **236**, 318 (2003).

[38] W. C. Thoburn, S. Legvold, and F. H. Spedding, Magnetic properties of terbium metal, Phys. Rev. **112**, 56 (1958).

[39] M. Mansuripur and M. Ruane, Mean-field analysis of amorphous rare earth-transition metal alloys for thermomagnetic recording, IEEE Trans. Magn. **22**, 33 (1986).

[40] J. Mendil, P Nieves, O. Chubykalo-Fesenko, J. Walowski, T. Santos, S. Pisana, and M. Münzenberg, Resolving the role of femtosecond heated electrons in ultrafast spin dynamics, Sci. Rep. **4**, 3980 (2014).

[41] P. Nieves and O. Chubykalo-Fesenko, Modeling of ultrafast heat- and field-assisted magnetization dynamics in FePt, Phys. Rev. Appl. **5**, 014006 (2016).

[42] M. S. El Hadri, M. Hehn, P. Pirro, C.-H. Lambert, G. Malinowski, E. E. Fullerton, and S. Mangin, Domain size criterion for the observation of all-optical helicity-dependent switching in magnetic thin films, Phys. Rev. B **94**, 064419 (2016).

[43] T. A. Ostler, U. Atxitia, and R. W. Chantrell (unpublished).

[44] I. Radu, C. Stamm, A. Eschenlohr, F. Radu, R. Abrudan, K. Vahaplar, T. Kachel, N. Pontius, R. Mitzner, K. Holldack, A. Föhlisch, T. A. Ostler, J. H. Mentink, R. F. L. Evans, R. W. Chantrell, A. Tsukamoto, A. Itoh, A. Kirilyuk, A. V. Kimel, and T. Rasing, Ultrafast and distinct spin dynamics in magnetic alloys, SPIN **05**, 1550004 (2015).