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Model for multishot all-thermal all-optical switching in ferromagnets

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All-optical magnetic switching (AOS) is a recently observed rich and puzzling phenomenon that offers promising technological applications. However, a fundamental understanding of the underlying mechanisms remains elusive. Here we present a model for multishot helicity-dependent AOS in ferromagnetic materials based on a purely heat-driven mechanism in the presence of magnetic circular dichroism (MCD). We predict that AOS should be possible with as little as 0.5% of MCD, after a minimum number of laser shots heat the sample close to the Curie temperature. Finally, we qualitatively reproduce the all-optically switched domain patterns observed experimentally by numerically simulating the result of multiple laser shots on an FePtC granular ferromagnetic film.

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The magnetic dynamics of a system triggered by an ultrashort optical pulse has been an exciting and yet unresolved problem in the magnetism community for the past 20 years. One of the most surprising results in the field was the discovery of the all-optical switching (AOS), the possibility of optically switching the magnetization of a magnetic material [1]. This was first observed in ferrimagnetic GdFeCo alloys [1], later in a variety of ferrimagnet alloys and synthetic ferrimagnets [2], and eventually in metallic ferromagnets [3]. It has been recently shown [4] that two different types of switching need to be considered: First, a single-shot helicity-independent AOS found in ferrimagnets such as GdFeCo [5], and second, a multishot helicity-dependent AOS found in TbCo and ferromagnets [4]. Although some models have been proposed that explain the switching in two-sublattice systems such as ferrimagnets [5–8], modeling for AOS in ferromagnets was lacking until very recently. To our knowledge, the only mechanism suggested so far for AOS in ferromagnets is a combination of heating and the inverse Faraday effect (IFE) [9,10]. The IFE corresponds to the generation of an effective magnetic field on the sample induced by the helicity of the light. However, IFE is still very difficult to characterize experimentally and in the cited works the amplitude of such fields is treated as a free parameter.

In this Rapid Communication, we present an alternative model that describes multishot helicity-dependent AOS in ferromagnetic materials based on a purely heat-driven mechanism. We first present the switching mechanism which is based on a combination of MCD and stochastic switching close to the Curie temperature T_C. This is followed by an in-depth description of the problem and of the way physical parameters are chosen. AOS is shown to be possible within a range of temperatures (i.e., laser fluences), for a large range of MCD values, but only after a certain number of laser shots. Finally, we reproduce previous AOS results by simulating the sweeping of the laser beam.

In our model, the mechanism driving the switching is a very simple and intuitive one: Whenever a laser heats a magnetic layer close to T_C, the stability of the magnetic state will be dramatically lowered as the anisotropy drops. For a circularly polarized beam, regions of the magnet with opposite magnetization will absorb different amounts of light due to MCD, resulting in hotter T_{hot} and cooler regions T_{cold}. The difference in temperatures will lead to a difference in magnetic stability. If $T_{\text{hot}} \approx T_c$ and the MCD is large enough, cool regions will remain stable, whereas hot regions will be prone to stochastic switching. Repeating the process (laser heating and cooling) multiple times will statistically lead the magnet to full switching.

In order to numerically test this idea, we represent the magnetic material by an array of N_x by N_y cells, grains, or macrospins [as in Fig. 1(a)], presenting a strong out of plane magnetic anisotropy of energy density K. For the sake of simplicity we ignore exchange and dipolar couplings between cells. These hypotheses will be discussed later in the text. This means that the magnetic state of the cell can be represented by a symmetric double well potential, as shown in Fig. 1(c), where the magnetization can be only in the states *up* or *down*. The characteristic hopping time for the magnetization from state a (*up*) to state b (*down*) is given by the Néel-Brown formula [11]:

$$\tau_{ab}(T) = \tau_0 \exp\left(\frac{E_{ab}(T)}{k_B T}\right), \tag{1}$$

where τ_0 is an attempt time typically estimated to be on the order of 0.1 ns [12], k_B is the Boltzmann constant, T is the temperature of the cell, and E_{ab} is the energy barrier that prevents the magnetization from switching. The barrier will have the temperature dependence $E_{ab}(T) = K(T)V$ [depicted in Fig. 1(b)] where V is the volume of the cell. A high anisotropy at room temperature T_0 leads to long-term stability of the magnetization. However, when heating the cells with a laser pulse (assuming a steplike heating profile of amplitude ΔT proportional to the laser fluence and duration t_{hot}) the probabilities that determine the final state (a or b) of the magnetization when starting in state a are given by [13]

$$P_{ab} = \frac{1}{2}\left[1 - \exp\left(-\frac{t_{\text{hot}}}{\tau_{ab}(T_0 + \Delta T)}\right)\right], \tag{2a}$$

$$P_{aa} = 1 - P_{ab}. \tag{2b}$$

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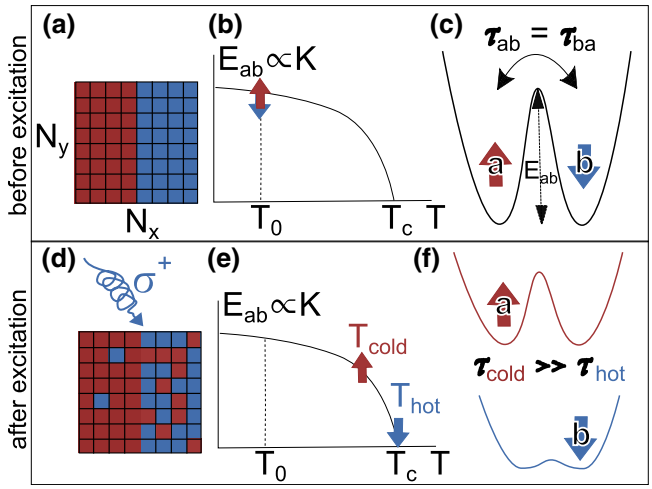

FIG. 1. The AOS model supposes an (a) N_x by N_y grid of cells, which present (b) temperature-dependent magnetic anisotropy energy barriers E_{ab}, resulting in (c) two stable possible states a (up) and b (down). As a (d) circularly polarized laser pulse arrives onto the grid, (e) cells in a and b states will absorb different amounts of energy due to MCD inducing a temperature distribution in the grid of hot and cold cells. This will lead to (f) different energy landscapes for the hot and cold cells, resulting mostly in stochastic switching of the hotter cells.

The probabilities are defined the same way when starting in state b, but the hopping time will be given by τ_{ba}. The probability function spans from 0 to $\frac{1}{2}$ as the energy barrier decreases from infinite to 0. Intuitively, it means that when the cell is heated close to T_C and the barriers disappear, the magnetization has no preferential direction.

If the laser pulse is right σ^+ (left σ^-) circularly polarized [14], b (a) states will absorb more heat due to MCD. This difference will result in hot and cold cells where, assuming a temperature-independent phonon heat capacity the temperatures are given by $T_{\text{hot/cold}} = T_0 + (1 \pm \frac{MCD}{2})\Delta T$ [Fig. 1(e)]. Because of this difference in temperatures, hot and cold cells will have different switching probabilities [Fig. 1(f)] leading to a higher number of reversals of the hot cells [Fig. 1(d)]. If we now heat the cells and allow them to cool back to T_0 N times, the cumulative probability for the magnetization to end in state b is given by (details in the Supplemental Material [15]),

$$P_B = \left(P_{ib} - \frac{P_{ab}}{P_{ab}+P_{ba}}\right)(1 - P_{ab} - P_{ba})^{N-1} + \frac{P_{ab}}{P_{ab}+P_{ba}}, \tag{3}$$

where the subscript i refers to the initial state. As N increases, the probability is given by the last term in Eq. (3) and when $P_{ab} \gg P_{ba}$ we find $P_B \approx 1$. Consequently, as long as enough heating cycles (i.e., laser pulses) are used and as long as there is a significant difference in the energy barriers for a and b cells, deterministic switching is expected. The barrier height difference originates in the difference in absorption due to MCD which leads to a different temperature rise for cells in states a and b. Because of this temperature difference and the strong [15] dK/dT close to T_C, E_{ab} will be very different for the two initial magnetization states.

Numerical simulations were conducted by considering an FePt-C-$L1_0$ granular film, for which AOS has been reported [3]. The typical size of grains is around 5 nm wide and 7 nm thick [16], where grains are separated by a 1-nm-thick C matrix [17]. This matrix ensures thermal isolation, as well as magnetic exchange isolation. We can therefore safely neglect the exchange interaction and assume that a nonhomogeneous temperature distribution can exist in the sample.

The temperature dependence of the magnetization M_S and K was extracted from Ref. [18], and corresponds to an $Fe_{50}Pt_{50}$ film. First, M_S was fitted with phenomenological equation [19] $M_{s0}[(T_c - T)/(T_c - T_0)]^\gamma$ where $M_{s0} = 1.15 \times 10^6$ A/m is the magnetization at $T_0 = 300$ K, $T_c = 775$ K, and $\gamma = 0.34$ is the phenomenological fitting exponent used for Fe [19]. Then K was fitted with [18,20] $K_0(M_S/M_{S0})^2$ where $K_0 = 4.5 \times 10^6$ J/m^3. The fits are shown in the Supplemental Material [15].

The MCD was calculated, for a wavelength $\lambda = 810$ nm, by using the nonmagnetic complex index of refraction $n = 3 + 4i$ and the complex nondiagonal term $\sigma_{xy} = -(1.4 + 1.7i) \times 10^{14}$ s^{-1} (cgs) of the optical conductivity tensor. These values were extracted from ellipsometry, Kerr rotation, and Kerr ellipticity measurements in Refs. [21,22] through the relations reported in Refs. [21,23]. Through Maxwell equations, the complex index of refraction for left and right circular polarized light $n_\pm$ are found to be [23] $n_\pm = \sqrt{n^2 \pm 4\pi\sigma_{xy}/\omega}$, where $\omega = 2\pi c/\lambda$ and c is the speed of light. Fresnel equations were then used to obtain the reflectances and absorptions A_+ and A_- for both helicities in the case of an infinitely thick film and normal incidence. Finally, the MCD was calculated as $2(A_+ - A_-)/(A_+ + A_-)$. An MCD of 5.8% is obtained for $Fe_{50}Pt_{50}$.

We first compute Eq. (3) as a function of the temperature, where the starting state a is heated more than b due to MCD. We begin with the calculated MCD = 5.8%, a cell volume of $5 \times 5 \times 7 = 175$ nm^3 [16], and $t_{\text{hot}} = 1$ ns. As shown in Fig. 2 for a single pulse ($N = 1$, black line) the probability

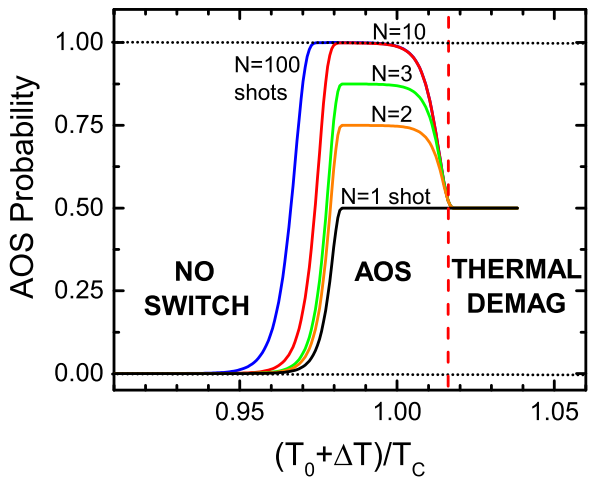

FIG. 2. All-optical switching probability as a function of the laser temperature increase ΔT for different numbers of shots N. Three different temperature regimes are observed. At low temperature no switching is possible due to strong anisotropy barriers. Close to T_C a certain amount of AOS occurs as the number of shots increases. With enough pulses full switching becomes possible. At higher temperatures the sample gets randomly demagnetized into a multidomain structure.

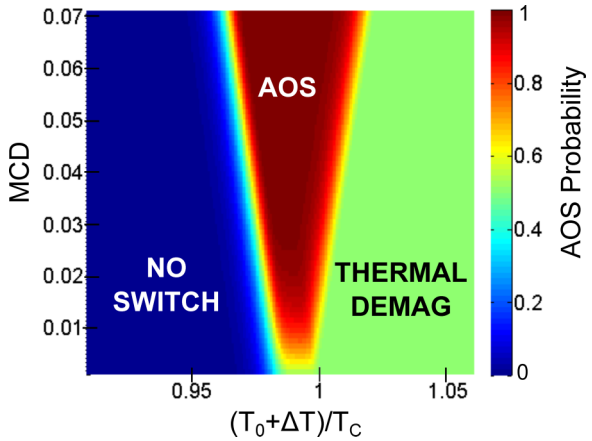

FIG. 3. AOS Probability as a function of temperature and MCD for $N = 10$ pulses.

of switching is 0 below a certain temperature threshold and 0.5 above. No full switching is thus possible with a single shot. As we increase the number of shots, a narrow range of temperatures around T_C results in a probability of switching that increases up to 1 eventually ensuring full switching. In this case the absorbed critical fluences for AOS will be around $F = \Delta T h C \approx 1.2\,\mathrm{mJ/cm^2}$, where $\Delta T = T_C - T_0$, $h = 7\,\mathrm{nm}$ is the thickness, and $C = 3.5 \times 10^6\,\mathrm{J/(m^3\,K)}$ is the heat capacity of FePt [24].

Since this model assumes a sudden steplike temperature increase in the sample, cooling dynamics are not taken into account. Cooling in the presence of strong dipolar fields would make a full switching process less probable. However, this argument is consistent with the observation [3] that only thin films, with a small magnetization volume and thus smaller dipolar fields, exhibit nearly full AOS. Thicker films always show some degree of demagnetization (multidomains), and full switching is not observed.

In this calculation, the ratio t_{hot}/τ_0 that acts as a prefactor in the exponential of Eq. (2a) was set equal to 10. This parameter varies for different heat dissipation in the sample, but mostly offsets the temperature range at which AOS is observed (see Supplemental Material [15] for details).

As shown in Fig. 3, the temperature window for AOS becomes larger as the MCD increases. However, even for a small MCD value of 0.5%, some AOS is still possible in a narrow range of temperatures.

Next, the lateral spatial heating profile was assumed to be Gaussian, according to the laser intensity profile. The $1/e^2$ radius was set to 115 cells and the temperature rise to $\Delta T = 600$ K [Fig. 4(a)] and $\Delta T = 500$ K [Fig. 4(b)]. As shown in Fig. 4(a), with white and black corresponding to opposite magnetizations, one shot on an initially saturated grid results in a circular demagnetized pattern. As the number of shots increases, an outer ring, within the temperature window for AOS, fully switches. The fact that the first laser shots only result in a demagnetized area agrees with the observations on Co/Pt from Ref. [4]. The final state, a ring domain with an inner demagnetized area, resembles strongly the AOS results in FePtC reported by Lambert *et al.* [3].

We next scan the Gaussian beam (temperature profile) at a speed of one cell/t_{rep} where t_{rep} is the laser repetition period,

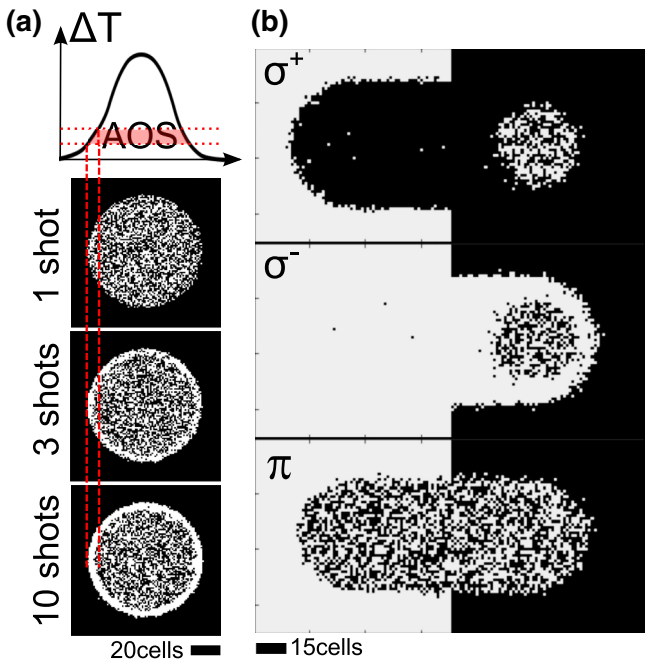

FIG. 4. Simulations of the magnetization state after assuming a Gaussian temperature increase induced by the laser intensity profile (profile details in text). (a) Simulation of successive shots with left circularly polarized (σ^-) light on an initially saturated black ("down") grid. After ten shots a fully switched white ring develops. (b) Simulations scanning the beam with left circular (σ^-), right circular (σ^+), and linearly polarized (π) laser shots at a ten shots/cell sweeping speed. Initially, the grid consisted of a left-half *up* (white) magnetization and right-half *down* (black) magnetization. Each helicity favors one magnetization direction, which is determined by the edge of the Gaussian profile, whereas the linearly polarized laser beam only demagnetizes the sample (see video in the Supplemental Material [15]).

and as shown in Fig. 4(b) (see video in the Supplemental Material [15]), we are able to write a magnetic domain by swiping the outer edge across the grid. Initially the grid was set so that the left half of the grid was *up* (white) and the right one *down* (black). Left circular (σ^-) and right circular (σ^+) polarized light causes *up* and *down* magnetizations, respectively, whereas linearly polarized (π) light only demagnetizes the sample and results in multidomain states. This is an expected behavior when considering the Gaussian profile of the laser intensity and the existence of a helicity-dependent AOS threshold [1]. In fact, this laser sweeping technique was used to reveal the AOS in ferromagnets such as FePtC by Lambert *et al.* [3] and lead to qualitatively similar magnetic domain patterns.

The switching time via this mechanism is obviously not ultrafast, in contrast to that found in ferrimagnets [6]. Since the film needs to be heated and cooled multiple times before observation of a full switching, the number of pulses needed for switching, cooling rates, and the laser repetition rate will define the switching time. However, such minimum switching time should also be fundamentally limited by the time scales of the thermal excitations (given by τ_0).

The suggested AOS mechanism should thus operate in materials with large MCD, small dipolar fields, limited in-

plane heat diffusion, and a strong temperature dependence of the anisotropy close to T_C. We note that this does not restrict the mechanism to ferromagnets; thin ferrimagnetic films (such as TbCo, GdFeCo) or even antiferromagntic materials are also strong candidates for this switching mechanism due to their small dipolar fields and significant MCD.

Finally we would like to discuss the validity of the model for ultrathin ferromagnetic films such as Pt/Co multilayers. These films do have significant MCD and exhibit low dipolar fields, however, they lack the granular structure that allows for magnetic and thermal isolation. Nevertheless, exchange interaction is not necessarily detrimental for the AOS. In such materials, we provide the following qualitative description: The first laser shot demagnetizes the sample. The magnetization spontaneously breaks into domains of various sizes. Under negligible dipolar fields, domain relaxation is dominated by the wall energy and pinning. The smallest domains will thus disappear because shrinking forces induced by the wall energy increase as the bubble diameter decreases [25], whereas larger ones (100–1000 nm wide) will remain pinned. These larger domains will then remain cool on the next laser shot, while the rest of the film will restart the same process over. Repetition of this mechanism will eventually finish when various large domains merge together, resulting in full AOS.

In summary, we have proposed a multishot all-thermal mechanism for helicity-dependent AOS in magnetic materials, which is based on temperature distributions induced by the MCD when heating the films close to T_C [26]. This mechanism could possibly coexist with other mechanisms such as the IFE [9,10]. We calculated the cumulative probability for AOS after a certain number of pulses, and numerically estimated it for the case of an FePtC granular film. The AOS window as a function of MCD, temperature, and the number of pulses was presented showing that even with as little as 0.5% of MCD, multishot switching should still be possible. Finally we simulated the resulting domain patterns after N laser shots, qualitatively reproducing various reported experimental results [3,4].

Note added. Recently, we discovered the independent work of Takahashi *et al.* [26] where they experimentally observe and study multishot AOS in FePt films. The authors fit the data assuming different switching probabilities, and even consider that the origin of such asymmetry could arise from MCD. Their observations strongly agree with our predictions.

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[14] Left, right circular and linear polarization are simulated by setting a positive, negative, and null MCD, respectively.

[15] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.94.020409 for a detailed calculation of Eq. (3), the M vs T for FePt, a brief discussion about the prefactor in Eq. (1), and a video showing the evolution of the magnetization as the laser beam is swept [Fig. 4(b)] is also available.

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