Influence of magnetic fields on ultrafast laser-induced switching dynamics in Co/Gd bilayers

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Recently it has been shown that not only GdFeCo alloys exhibit single-pulse helicity-independent all-optical switching, but that this effect is also seen in Co/Gd bilayers. However, there have been no reports on the explicit time dynamics of the switching process in these bilayers as of yet. Furthermore, time-resolved measurements of switching of other materials are typically done with a constant applied field to reset the magnetization between consecutive pulses and thus ensure repeatable behavior. In this paper we experimentally resolve the explicit dynamics of the switching process in Co/Gd, and the influence of applied magnetic fields on the switching process. We observe that after a switch within several picoseconds, the magnetization switches back at a time scale of hundreds of picoseconds. This backswitch includes a strong dependence on the magnetic field strength even at subtesla fields, significantly smaller than the exchange fields that govern the switching dynamics. This surprising behavior is explained by a combination of longitudinal switching (on a picosecond timescale), precessional switching (on a nanosecond time scale), and domain-wall motion (on a timescale of 10 ns and beyond). We discuss these different switching regimes and their relative importance using simple model calculations.

I. INTRODUCTION

All-optical switching (AOS) of the magnetization is a phenomenon where the direction of the magnetization in a material can be switched using laser pulses [1–6]. It has the potential to be used in new types of data storage devices, since optically writing magnetic bits provides unprecedented speeds and high energy efficiency. Discovered in 2007, single-pulse helicity-independent all-optical switching (HI-AOS) was originally only found in GdFeCo alloys [1]. It is generally believed that the antiferromagnetic coupling between the two spin sublattices (Gd and FeCo) is essential for AOS to appear. Upon excitation with a laser pulse, both subsystems demagnetize on different time scales [7]. The transfer of angular momentum between the two sublattices, mediated by exchange scattering, is thought to be the driving force behind the switching [8]. Recently, it has been shown that AOS is also possible in Co/Gd bilayers, which is extremely interesting from an application point of view as it provides opportunities for interface engineering [9,10].

However, there have been no reports on the explicit time-resolved dynamics of the switching process in Co/Gd bilayers, raising the question whether switching in Co/Gd bilayers happens on the same ultrafast ps time scale as with the CoGd and GdFeCo alloys. Furthermore, most experiments on the dynamics of the switching process in AOS with time-resolved magneto-optic Kerr effect (TR-MOKE) or similar techniques have been performed using a constant applied magnetic field [7,11]. This field is used to return the magnetization to its original direction between successive laser

pulses. If the applied magnetic field has an influence on the measured dynamics, it is important to keep this in mind when analyzing time-resolved AOS experiments. Consequently, we want to answer a second question; how big is the influence of this field on the dynamics, and on what time scales does this influence become of relevance.

In this paper we will show a TR-MOKE method that can be used to measure the switching dynamics without applying a magnetic field. In Co/Gd (and GdFeCo) the magnetization switches via a highly stable toggle mechanism, alternating between up and down. A constant applied field can be used to return the magnetization to its original direction between successive laser pulses, but in our method we detect every other laser pulse, thereby measuring either the up-down or the down-up switch without the need for an applied magnetic field. This is confirmed by the demonstration of switching for at least 10^8 pulses at a repetition rate of 100 kHz. In these time-resolved measurements we observe a similar ultrafast switching response in Co/Gd bilayers as in GdFeCo alloys. After that, we report on the field-dependence of the switching process, where a constant field of 0 to 400 mT is applied in the initial direction of the magnetization. We find that the initial switch and recovery to about 50% of the saturation magnetization (in the opposite direction) occurs in approximately 10 to 20 ps and is independent of the applied field. However, from that moment onwards the dynamics strongly depend on the applied field, and a field of several 100 mT suffices to obtain a backswitch within a few 100 ps. This is remarkable, as the exchange fields that govern the switching process are orders of magnitude higher than the applied fields that we use. We will discuss the various mechanisms that can play a role in this backswitch, namely longitudinal switching (described by a layered-microscopic three-temperature
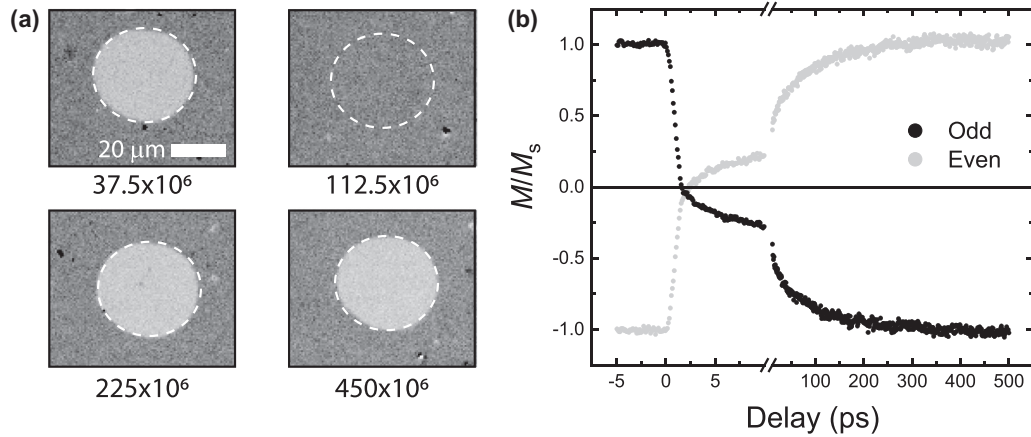
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FIG. 1. (a) Kerr microscope images of AOS in Co/Gd. Dark and light contrast corresponds to up and down magnetization, the dashed line indicates the region where the local fluence is high enough to reverse the magnetization. A switched domain is observed depending on whether the total number of pump pulses is even or odd. The labels indicate the (approximate) number of pulses for each image. The measurements are done with a repetition rate of 125 kHz. (b) Field-free time-resolved measurement of AOS in Co/Gd, by measuring only half the pulses, at 100 kHz. Using only the odd or even pulses corresponds to measuring the up-down or down-up switch, respectively. Note the axis break on the x axis.

model (M3TM) [12]), precessional switching [described by a Landau-Lifschitz-Gilbert (LLG) model] and switching via domain-wall motion, and we will examine the importance of these processes in the backswitch. We conclude that the backswitch is dominated by precessional switching, although not all features of the switching process can be explained by precessional switching alone.

II. METHODS

The measurements are performed on SiB//Ta (4 nm)/Pt (4 nm)/Co (1 nm)/Gd (3 nm)/Pt (4 nm) samples, which are grown by DC magnetron sputter deposition at room temperature, with a base pressure of $10^{-8} - 10^{-9}$ mbar. The Co and Gd are coupled antiferromagnetically, with roughly 0.5 nm of the Gd magnetized [9]. This proximity-induced Gd magnetization is in the opposite direction of the Co magnetization. Hysteresis loops show square loops [9], confirming a perpendicular magnetic anisotropy, with coercive fields of about 20 mT. The magneto-optic signal in our experiments is a linear combination of contributions by the Co and Gd subsystems, but is generally assumed to be dominated by Co in our wavelength range [13–15].

The time-resolved experiments are carried out with a Spectra-Physics Spirit-NOPA laser system. The pulse length at sample position is ∼100 fs, with a central wavelength of 700 nm at a repetition rate of 100 kHz. A high-aperture laser objective is used to focus the pump and probe pulses on the sample to a size (full width at half maximum) of ∼80 μm for the pump pulses and ∼30 μm for the probe pulses. In order to measure the switching dynamics without an applied field we want to measure only the even or odd pulses. This is achieved by adding a transistor between the photodetector and the lock-in amplifier. By synchronizing the signal applied to the base of the transistor with the laser we are able to block exactly half of the pulses in the switching process. A

more detailed explanation about the setup can be found in Supplemental Material Note I [16].

In order to investigate the robustness of the switching process we expose the sample to laser pulses at 125 kHz for increasing amounts of time. The result can be seen in Fig. 1(a), confirming that we achieve robust switching for up to 1 h, corresponding to 4.5×10^8 pulses. The labels indicate the (approximate) number of pulses for each image, resulting in a switched domain depending on whether the total number of pulses is even or odd. For too high laser fluences the combination of a relatively high repetition rate and the long exposure time causes damage to the sample, either by annealing the stack or by causing structural damage in the stack (not shown here). In our experiments, care is taken to avoid this damage.

III. FIELD-FREE SWITCHING

In Fig. 1(b) we show the TR-MOKE measurement on the switching dynamics in Co/Gd without applying a magnetic field, selecting either even or odd pulses. Time traces for even and odd pulses are exactly opposite, confirming the highly deterministic toggle switching. We are able to switch between measuring the even and odd pulses by changing the phase of the signal sent to the base of the transistor. In Supplemental Material Note II [16] we confirm that the measurements with transistor do not introduce any artifacts in the measured dynamics. As can be seen left of the axis break in Fig. 1(b), within several ps switching occurs, which is similar to the ultrafast switching seen in GdCo and GdFeCo alloys [11]. This answers our first question, i.e., switching in Co/Gd bilayers occurs at the same time scale as in GdFeCo alloys. On the right side of the axis break, after about 200 ps we see a complete recovery of the magnetization in the switched direction due to the cooling down of the system. We note that this recovery is due to the cooling
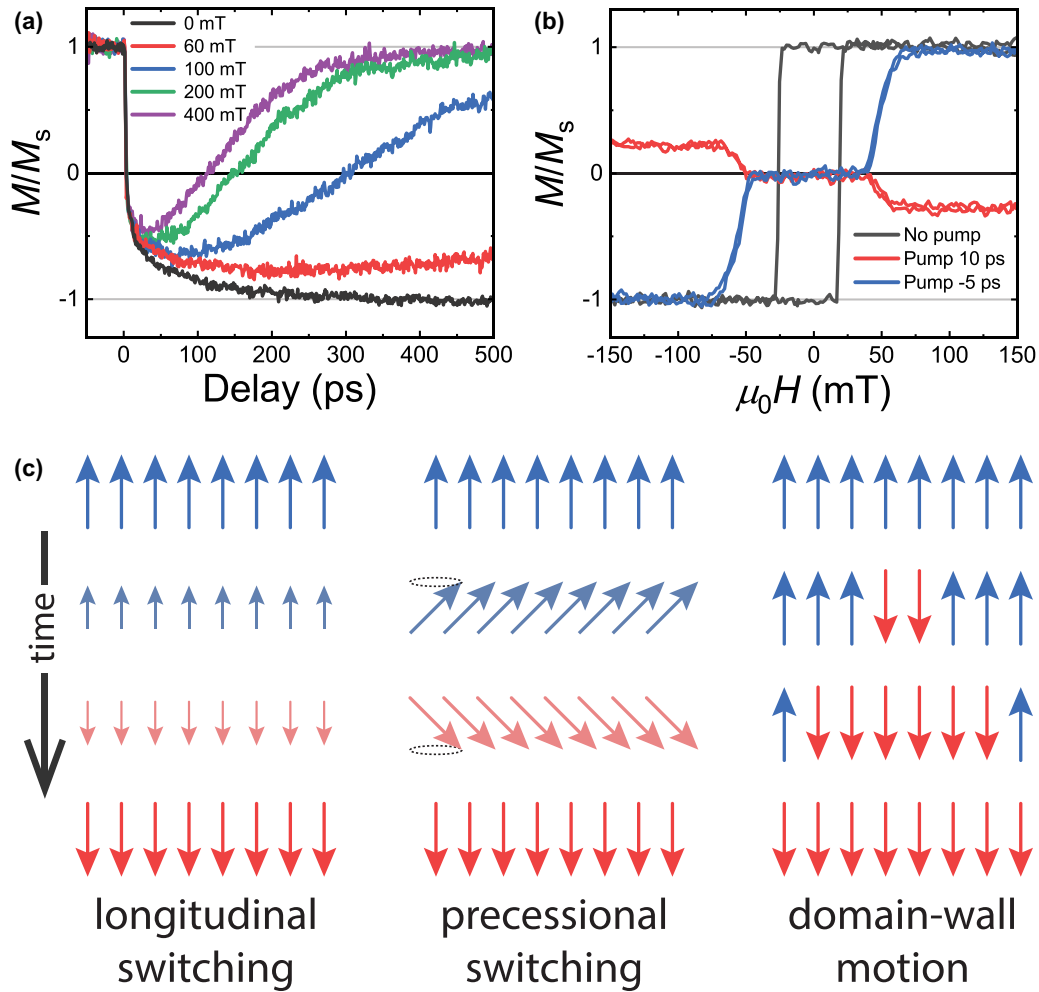
FIG. 2. (a) Time-resolved measurements of AOS with an applied magnetic field. The constant field is applied in the positive *z* direction. (b) Hysteresis loops with and without pump. The black line shows the hysteresis loop without pump, the red and blue lines show the hysteresis loops with pump and the delay fixed at −5 and 10 ps, respectively. (c) Schematic depiction of the three switching mechanisms examined in this paper: longitudinal switching, precessional switching, and switching via domain-wall motion.

down of the film, governed by the thermal diffusion into the substrate.

IV. FIELD-DEPENDENT SWITCHING

After having resolved the field-free switching, we turn towards the influence of a constant magnetic field, applied in the out-of-plane direction, on the switching dynamics. For strong enough fields, this will make sure that the magnetization switches back to the original magnetization direction between two successive pump pulses. For this reason this method is often used in pump-probe experiments. The influence of the field on the switching dynamics up to 500 ps can be seen in Fig. 2(a). We indeed observe the magnetization reversing back to the original direction due to the applied field, for fields larger than 100 mT even within the measured 500 ps. This answers the second question we posed: even at a sub-ns time scale the dynamics are indeed dependent on the magnetic field. For fields of several hundred mT we observe a deviation with the zero-field measurement already after tens of ps, while within hundreds of ps the magnetization switches back. Such behavior should be considered surprising, realizing that the

AOS process is governed by exchange fields of the order of 1000 T, more than three orders of magnitude larger than the applied field. The out-of-plane hysteresis loops visible in Fig. 2(b) give some more insight in the fields and time scales related to the magnetization reversal due to the applied magnetic field. In black a regular hysteresis loop is visible, measured without the pump pulses present, so no AOS happens in this case. The loops in blue and red are measured with pump pulses present (so AOS does happen in this case) at two different delays between the pump and probe. In blue the hysteresis loop is visible with the delay fixed at 10 ps, at which the magnetization is (partially) switched. This can be seen in the hysteresis loop and also in Fig. 1(b), both showing $M/M_s \approx 0.25$ after 10 ps. In these hysteresis loops we also see that fields up to 150 mT do not influence the dynamics on this time scale. In the red loop the delay is fixed at −5 ps, which corresponds to the probe pulse arriving about 10 μs after the pump pulse. For small fields the signal is zero, as the magnetization toggles between up and down, resulting in no net signal. Above about 55 mT the magnetization is at the same level as without pump, showing that the magnetization has fully returned to the initial direction after
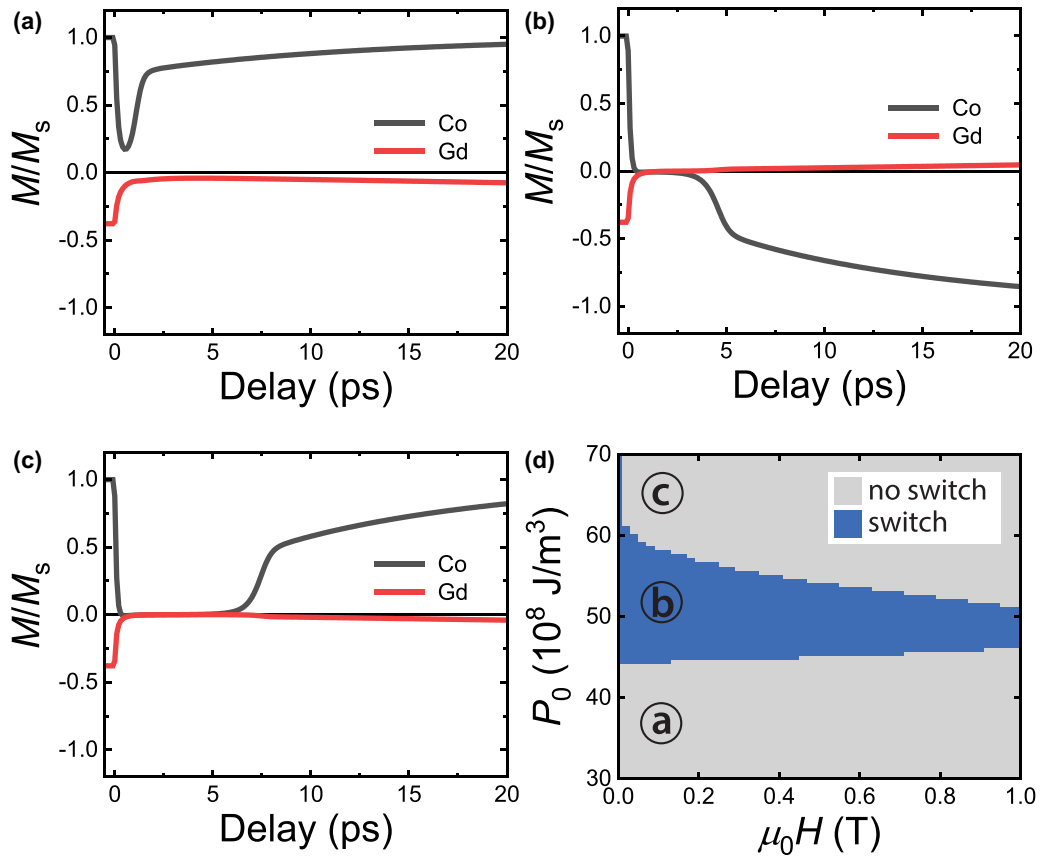
FIG. 3. M3TM calculations. (a) m_z after laser pulse excitation with a fluence below the threshold fluence. (b) m_z after excitation with a laser pulse above the threshold fluence. (c) m_z after excitation with a laser pulse above the threshold fluence and a magnetic field in the positive z direction strong enough to result in no switch. (d) Phase plot of the magnetization direction after 100 ps as a function of applied field and the laser fluence. The three regions correspond to the traces shown in (a), (b), and (c).

the switch. For this reason, a field of 55 mT is the minimum field for which we are able to measure the dynamics of AOS at 100 kHz.

The question arises as to what the driving force behind this reversal and successive return at suprisingly short time scales is. We will discuss three mechanisms, as schematically depicted in Fig. 2(c). First we address longitudinal switching, the driving mechanism behind AOS. We will model this process using a modified M3TM [12], as will be discussed in the next section. We conjecture that for time scales larger than 20 ps precessional switching, describing the switching due to applied torques on the magnetization, may play a role. This will be described using a modified LLG model, which will be introduced in Sec. IV B. Finally, we will discuss the role of field-driven domain-wall motion, which occurs on even longer time scales and smaller fields, and whereby the switched domain will collapse even if the two other mechanisms are too weak.

A. Longitudinal switching (M3TM)

The first mechanism we discuss is longitudinal switching, the process that is responsible for AOS in the first place. Typically, this works on very short timescales (<10 ps), but as can be seen in Fig. 2(a) a deviation between the experimental traces at different magnetic fields occurs on short time

scales already. The relevant field strengths in the switching process are the exchange fields, orders of magnitude larger than the fields that are applied in our experiment. The question is therefore whether the relatively small applied fields can significantly influence the switching process and could drive the backswitch via longitudinal switching, thereby explaining the dependence on the applied field in our experiments.

To simulate the switching process we use a layered-M3TM [12,17], which models each atomic layer as a separate system, interacting with neighboring layers via exchange scattering. In Supplemental Material Note III A [16] more details about this model can be found. In brief, we use five atomic layers of Co and three atomic layers of Gd, roughly corresponding to 1 nm of Co and 1 nm of Gd. The ground state magnetization profile of the Gd decays exponentially, and adding more Gd layers would not significantly change the resluts. A Gaussian laser pulse is used to excite the system, after which the magnetization dynamics are calculated in a layer-resolved fashion. In Fig. 3(a) the result of such a simulation is depicted for a relatively low laser energy, with the black and red line indicating the average Co and Gd magnetization, respectively. In this case, the laser fluence is not high enough for the electron temperature to reach the Curie temperature. Thus, after partial demagnetization the magnetization of both layers returns back to the initial direction, and there is no AOS. Above a certain switching threshold AOS is observed, as shown in Fig. 3(b). A

transient ferromagnetic state at a very low magnitude is visible as a plateau for several ps, after which the Gd magnetization crosses zero as well, which creates a switched state. In both these cases [Figs. 3(a) and 3(b)] no applied field is used. When introducing an applied field, aligned with the original Co magnetization, a third possibility appears. For the right combination of fluence and field strength, the magnetization is almost fully quenched, but returns to its original direction instead of switching, which can be seen in Fig. 3(c). This applied field-driven backswitch is a consequence of the long duration of the plateau that lasts for several ps. In this state with almost fully quenched magnetization, a relatively small magnetic field is enough to prevent the switch and cause the magnetization to go back to its original direction. In Fig. 3(d) a phase plot is visible where we show how the combination of laser fluence and applied magnetic field influences the switching behavior. The switched and nonswitched regions in the phase plot are determined by tracking the direction of the Co magnetization after 100 ps and comparing that with the original direction. For laser fluences below the threshold fluence no switching happens, as visible in the bottom grey area (a). For increasing magnetic fields, we see a slight increase in the threshold fluence. Above this area the region where switching occurs is visible (b). For zero field these are the only two possibilities, but for a finite field a third region (c) exists, which corresponds to the magnetization returning to its original direction. From the phase plot it is clear that this effect is a combination of high fluence and large magnetic field. This second threshold for backswitching strongly depends on the applied magnetic field, with a higher fluence requiring a smaller magnetic field to achieve this. This can be understood from the strong reduction of effective exchange field in the strongly quenched magnetic state, resulting in applied fields and exchange fields of similar magnitudes.

Although this behavior looks similar to what we see in our experiment, where an external field can result in the magnetization returning to the original direction, the mechanism at play does not appear to be the same. The pronounced plateau around zero magnetization— persistent for more than 5 ps in these calculations— is not visible in our experiments, both with and without an external applied field. We verified that this plateau is the crucial driving force for the backswitch mechanism in the layered-M3TM. We conjecture that the occurence of such a plateau is an artefact of the M3TM, as this model does not take into account thermal or spatial fluctuations. In models that do take into account these fluctuations, such as atomistic LLG calculations, this plateau is not visible, more in line with the experiment. As a consequence, we expect the strong field dependence not to be visible in such models. We do stress that there might be a mechanism of longitudinal switching playing a role in the switching back, although one that is not captured by our simple model.

B. Precessional switching (LLG)

The second mechanism that we discuss is precessional switching, which describes the behavior of the magnetization under the influence of torques applied to it, in our case due to a combination of the externally applied field and the local anisotropy fields. To describe this behavior we use an LLG

model, where we explicitly take into account the change in magnitude of the magnetization because of the AOS. Thus, we write $|\mathbf{M}(t)| = \chi(t)M_s$, where we assume that $\chi(t)$ is not affected by the magnetic field. Substituting this relation into the LLG equation and dividing out $\chi(t)$, this results in the following equation for the transverse dynamics:

$$\frac{d\mathbf{m}}{dt} = -\frac{\gamma\mu_0}{1+\alpha^2}(\mathbf{m} \times \mathbf{H}_{\text{eff}} + \alpha\chi(t)\mathbf{m} \times \mathbf{m} \times \mathbf{H}_{\text{eff}}), \quad (1)$$

with $\mathbf{m}$ as the normalized magnetization, γ as the gyromagnetic ratio, μ_0 as the vacuum permeability, α as the Gilbert damping constant, and $\mathbf{H}_{\text{eff}}$ as the effective field—comprised of the external field and the anisotropy field. Furthermore, $\chi(t)$ is the time-dependent magnitude of the magnetization given by a double exponential fit to the data from Fig. 1(b), which starts at a positive value and becomes negative after the switch. In Supplemental Material Note III B [16] a more detailed explanation of the model is given.

In our simulations we define the z axis as the out-of-plane (OOP) axis. The initial magnetization is in the z direction and the magnetic field in the xz plane, making an angle θ with the z axis. This is done to incorporate the effect of a possible small misalignment of the magnetic field with the easy axis of the sample in the experiments, as well as the fact that due to thermal fluctuations on a microscopic scale there will be a finite angle between the local magnetization and the easy axis. Rather than describing an inhomogeneous system with a spatially (and temporally) fluctuating local magnetization, we limit ourselves to a single macrospin with a fixed orientation with respect to the applied field to capture the basic physics. The parameters used in these calculations are realistic values for a Co/Gd bilayer system and can be found in Supplemental Material Note III B.

We first let the magnetization relax to an equilibrium state, after which at $t = 0$ the magnetization starts to switch to the opposite direction via longitudinal dynamics described by $\chi(t)$. In Fig. 4(a) the magnetization in the z direction from these calculations is shown for various magnetic fields, assuming $\theta = 0.05$ rad. For the first 200 ps no significant influence of the applied field is visible. In all cases there is a demagnetization, followed by a reversal of the magnetization direction and a remagnetization in the opposite state. On longer time scales the magnetization reverses to the initial direction; more quickly for the higher applied fields. Qualitatively, this corresponds well with the experiments [Fig. 2(a)], as the magnetization reverses back to the original direction within 500 ps. However, several differences are noteworthy. First, the fields needed to establish a sub-ns backswitch in the calculations are a factor two to three higher than the fields in the experiment. This might be explained by our neglect of thermal fluctuations. Although in the model the angle θ between easy axis and magnetic field direction plays a similar role, this should be considered a severe approximation. Furthermore, we notice that in our calculations up to 200 ps no field dependence is seen, in contrast to our experiments. Moreover, the reversal happens faster in the calculations than in the experiments. Apart from the already mentioned thermal fluctuations, another factor that might explain these differences is the assumption of a single macrospin, instead of the two coupled subsystems that are present in the bilayer. To explore
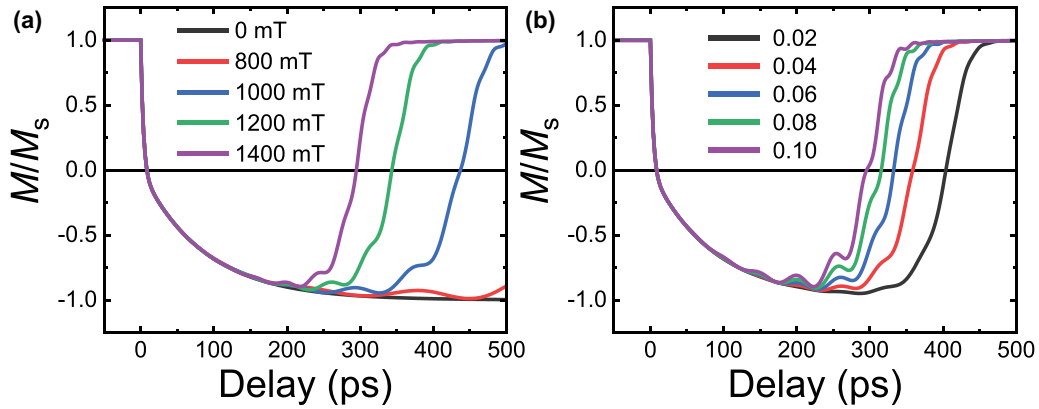
FIG. 4. LLG calculations. (a) z magnetization as a function of delay for various magnetic fields, pointing in the positive z direction. The angle between the applied field and the z axis is $\theta = 0.05$ rad. (b) z magnetization as a function of delay for various values of θ. The applied field is $\mu_0 H = 1200$ mT.

other reasons for the quantitative mismatch between simulations and experiments, we have looked into the influence of the damping α, the effective anisotropy K_{eff} and the saturation magnetization M_s as well, but found no qualitative difference in the switching process. Details of these calculations can be found in Supplemental Material Note IV [16].

Note that the oscillations that we observe in Fig. 4 are a trivial consequence of our macro-spin approach. They emerge as a result of the angle between the easy axis and **H**, mainly visible for bigger angles as can be seen in Fig. 4(b). Therefore we do not expect these oscillations to occur in experiments.

To conclude, the dynamics of the backswitch within 500 ps are thus qualitatively well explained by our simple LLG model, but some detailed features cannot be fully reproduced, mainly with regards to the behavior within 200 ps. While this is partly due to simplifications made in our model, it is possible that other effects might play a role as well. For example, the longitudinal time dependence of the magnetization is assumed to be independent of the magnetic field. A more realistic approach that takes into account the field dependence of $\chi(t)$ may be needed for a better agreement with the experiments.

C. Domain-wall motion

Lastly we describe domain-wall (DW) motion as a switching mechanism. If the applied field is in the same direction as the magnetization outside of the switched region it will cause the switched domain to shrink and eventually collapse. This effectively results in a backswitch of the domain, similar to precessional switching. However, this process takes place on a time scale an order of magnitude longer. We first discuss the effect of DW motion on the dynamics within 500 ps, as seen in Fig. 2(a), and after that the effect on longer time scales, as can be seen in Fig. 2(b). Assuming a switched domain with a radius of 15 μm and a domain-wall velocity of 200 ms^{-1} at 60 mT[18] we find that it takes 75 ns for the domain to disappear, and thus that within 500 ps the DW travels 0.1 μm. Relating this to the measurements from Fig. 2(a), it is safe to conclude that the dynamics in this measurement are unaffected by the influence of DW motion via the edge of the switched domain, as the domains have a diameter of ∼30 μm.

However, this assumes a single nucleation site within the switched region. If more backswitched domains are formed, that would increase the effect of DW motion on the total switched area. However, only for a large number of nucleation sites is this expected to be significant, as these individual DWs still travel only 0.1 μm in 500 ps.

Although we excluded DW motion to be of relevance for the dynamics within 500 ps, it can play a role in ensuring the magnetization reverses to its original direction between two successive pulses. This can be seen in the hysteresis loop for −5 ps in Fig. 2(b), which shows the magnetization just before the next pulse arrives. The DW velocities mentioned above are certainly high enough to achieve a collapse within 10 μs (the time between two successive pulses), although we do not have enough information to assess the exact magnitude of the DW motion in this regime. In Fig. 5(a) the process through which DW motion can reverse is shown, with the dark regions corresponding to the up magnetization and the light regions corresponding to the down magnetization. Time goes from left to right, with first a switched domain (I), then the domain shrinking due to domain-wall motion (II) and fully disappearing (III) before the next pulse arrives (IV).

For this process a certain field is needed for which the magnetization is fully reversed before the next pump pulse arrives. From Fig. 2(b) we estimate that the minimum applied field is approximately 60 mT, corresponding to a DW velocity of 1.5 ms^{-1}. If the field is smaller than that, a more intricate progression may appear. This is represented in Fig. 5(b). Again we see a switched domain (I), then the domain shrinking due to the applied field (II and III). However, now the domain is not fully reversed, and a ring pattern is expected to appear after the next pulse (IV). This ring pattern will again shrink, now from both sides (V). Due to this the domain can be fully reversed before the next pulse arrives (VI), and the cycle starts from the beginning. Probing a corresponding doubling of the periodicity would provide direct evidence of this DW motion driven dynamics, but is beyond the scope of our present paper.

V. CONCLUSIONS

To conclude, in this paper we showed field-free measurements of the switching dynamics in Co/Gd bilayers, by using
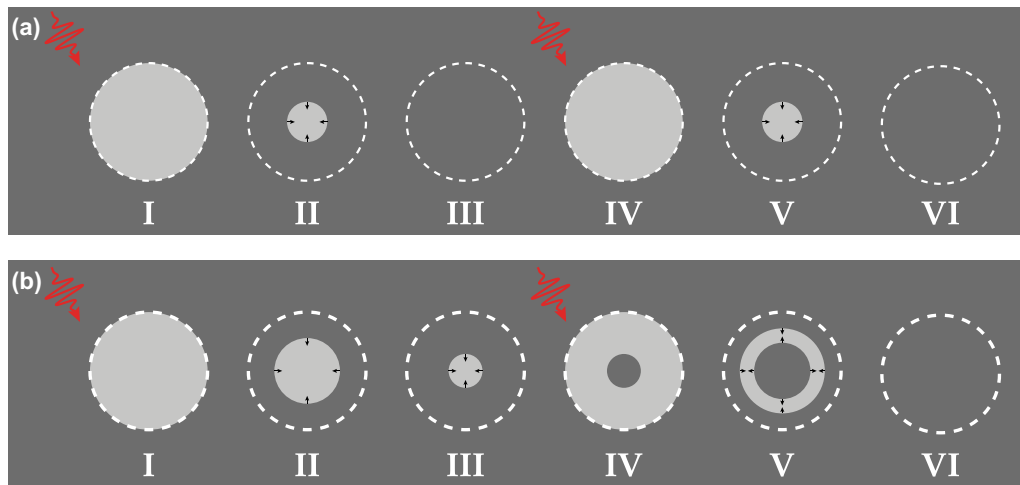
FIG. 5. Schematic depiction of the cycles that can appear in the switching process due to domain-wall motion and AOS, with time going from left to right. The black arrows indicate the direction of DW motion due to the applied field. (a) Cycle for an applied field strong enough to fully reverse the magnetization between two pulses. (b) Cycle for a smaller field, resulting in a longer cycle.

the toggle-switching behavior of HI-AOS and measuring every other pulse. We confirm that AOS in Co/Gd happens at the same ultrafast rate as in the alloys CoGd and GdFeCo. When applying an external field, we observe a strong field dependence within 500 ps, visible as a switch back to the initial magnetization direction. Discussing various switching mechanisms (longitudinal switching, precessional switching, DW motion) we conclude that the fast backswitch is dominated by precessional switching, but that this does not fully explain the observed behavior, especially on time scales below 50 ps. Furthermore, we conjecture that at smaller fields a collapse of the switched domain will be driven by DW motion. We predict the possibility to observe a doubling of the periodicity of the dynamics in this regime.

Finally, we expect our work to trigger further research aimed at more quantitative explanation of the magnetic field dependent behavior. Independent of the underlying mechanism, it will be important to keep in mind all these effects when analyzing time-resolved AOS experiments in the presence of an applied field.

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[16] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.105.014429 for (I) a description of the setup; (II) a comparison between measurements with and without transistor; (III) details about the layered-M3TM and LLG model; and (IV) effects of several material parameters on magnetization reversal in the LLG model.

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