

Athermal all-optical femtosecond magnetization reversal in GdFeCo

J. Hohlfeld,^{1,a)} C. D. Stanciu,^{1,2,b)} and A. Rebei^{1,c)}

¹Seagate Research Center, Seagate Technologies, Pittsburgh, Pennsylvania 15222, USA

²Institute for Molecules and Materials, Radboud University Nijmegen, Toernooiveld 1, 6525 ED Nijmegen, The Netherlands

(Received 5 March 2009; accepted 16 March 2009; published online 14 April 2009)

Magnetization reversal in GdFeCo by circularly polarized light is shown to occur at the femtosecond time scale. In contrast to the well-known laser-assisted magnetization reversal based on the laser heating, we here demonstrate that this femtosecond all-optical magnetization reversal is more efficient at lower temperatures. Both of these findings make all-optical recording on rare-earth transition metal alloys a promising technique for stable magnetic storage with high data rates. © 2009 American Institute of Physics. [DOI: 10.1063/1.3119313]

Using 40 fs circularly polarized laser pulses, it has been recently demonstrated that laser alone can be used to control the direction of the magnetization.¹ It was shown that the reversal of the magnetization can be controlled by the helicity of the light and no external magnetic field is required. This magnetization reversal mechanism was originally understood as the combined result of femtosecond laser heating of the magnetic system to just below the Curie point and circularly polarized light simultaneously acting as a magnetic field. On the other hand, it has been argued recently that this magnetization reversal is completely *athermal*, and therefore does not require heating near the Curie temperature.² More specifically, it was proposed that femtosecond athermal switching is possible in rare-earth doped transition metals via an optical Barnett-like effect³ in the presence of strong fluctuations and dissipation due to strong laser-enhanced spin-orbit coupling [such as recently demonstrated in ferromagnetic Ni (Ref. 4)]. Nevertheless, the mechanisms responsible for the all-optical *permanent* magnetization reversal in metals are currently a matter of debate. Besides this, another open question is related to the all-optical switching reversal speed. In Ref. 1, it is shown that each circularly polarized 40 fs laser pulse leads to the formation of a permanent magnetic domain. But what is the timescale of this reversal?

In this letter, using a time resolved pump-probe setup, we investigate the reversal time of all-optical switching. It is experimentally demonstrated that the reversal occurs in the subpicosecond regime. In agreement with a very recent observation,⁴ this subpicosecond switching rate indicates that relaxation effects induced by optical excitations are much larger than those derived from ferromagnetic resonance (FMR) measurements. In addition, by investigating the temperature dependence of the all-optical switching we show that this process is taking place even at low temperatures. More specifically, the lower the temperatures, the smaller the laser fluence required for the switching. This observation demonstrates the pure athermal origin of the reversal mechanism. The time-resolved experiments were performed in Pittsburgh, employing relatively long laser pulses, of ≈ 500 fs duration, while the temperature dependent measurements were carried out in Nijmegen using 40 fs laser

pulses. The metallic magnet studied here was an amorphous $\text{Gd}_{22}\text{Fe}_{74.6}\text{Co}_{3.4}$ thin film as in Ref. 1.

We first discuss the time-resolved measurements. The pump-probe pulses use a two-color scheme with a wavelength of $\lambda=800$ nm pump and $\lambda=400$ nm probe pulses. The pump pulses of 100 μJ energy are focused at normal incidence onto the sample, via a 700 mm lens. The collinear probe pulses are attenuated to 1 μJ to avoid significant self-action, and are focused with the help of a 100 mm lens. The diameter of the focused pump and probe beams was estimated to be of the order of 500 and 50 μm , respectively. The Kerr rotation of the probe pulses was recorded by a standard two diode scheme⁵ and lock-in detection using the 500 Hz repetition rate of the laser system as a reference frequency. The sample is mounted on a fast rotation stage, which ensures that every pump-probe pulse pair is exciting/probing a fresh spot initialized by a nearby magnet. The pump-pulse duration was determined by comparing measured and calculated second-order intensity correlation functions to be ≈ 500 fs. Using relatively long pump pulses, the optically excited electrons have lots of time to thermalize among each other and even to transfer a significant amount of energy to the lattice while the pump pulse is on. Hence, we observe a significant reduction of the magnetization magnitude during the reversal process (Fig. 1).

In order to separate switching dynamics from purely temperature induced changes in the magnitude of the magnetization, we use a procedure described in Ref. 6. Here, we use the normalized out-of-plane component of the magnetization, m_z , derived from the ratio of the Kerr signals measured for both pump helicities, i.e., for the switching and nonswitching cases, to extract the switching dynamics. It is clear from the time dependence of m_z , shown in the inset of Fig. 1, that the switching is very fast and occurs within 1 ps. For our laser pulse of width 500 fs, we estimate that the switching is complete within 700 fs. The switching is therefore too fast to follow a precessional path. Indeed, the model discussed in Ref. 2 shows that precessional dynamics is not important for femtosecond magnetization reversal. Figure 1 also shows that our measurements yield a minimum value of $m_z = -0.5$ instead of -1.0 , thereby indicating that on average only 75% of the pump pulses lead to a reversed state. Because the all-optical switching is strongly sensitive to a change in the pump fluence,¹ a complete 100% reversal requires a very fine tuning of the laser fluence, which was

^{a)}Electronic mail: julius.hohlfeld@seagate.com.

^{b)}Electronic mail: cd.stanciu@gmail.com.

^{c)}Electronic mail: arebei@mailaps.org.

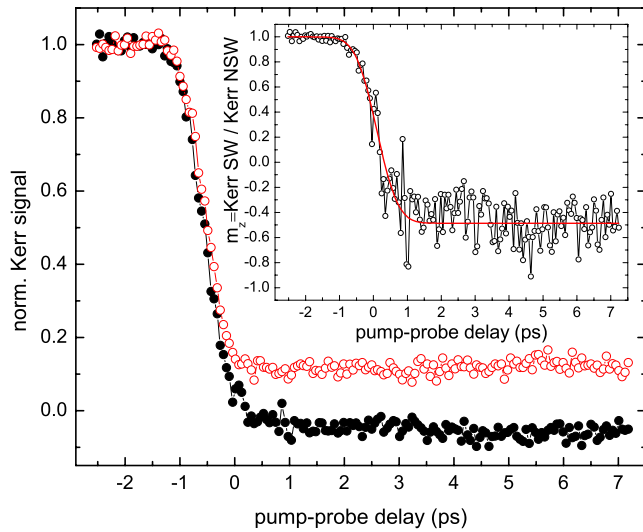


FIG. 1. (Color online) Transient magnetization dynamics induced by circularly polarized pump pulses with helicity favoring the initial orientation of the magnetization (NSW, open symbols) and leading to magnetization reversal (SW, closed symbols). The inset shows the resulting dynamics of the normalized out-of-plane component of the magnetization, m_z , obtained by dividing the transient for the switching case by the one for the nonswitching configuration. The line is to guide the eye and represents an error function of 700 fs width.

difficult to attain in our experiments. Shorter laser pulses should yield stable switching over a wider fluence range and be able to diminish the effect of heat on the magnetization.

To obtain femtosecond magnetization reversal by circularly polarized light, three main requirements must be fulfilled: (1) an ultrafast channel for angular momentum exchange, between the spins and another degree of freedom, such as lattice; (2) a light-induced switching mechanism, where the magnetization direction is controlled by the light helicity; (3) a mechanism that, after switching, maintains information about light helicity in spite of the decoherence effects that takes place in metallic magnets on the femtosecond time scale. We will now briefly discuss each of these mechanisms:

- (1) We observe an all-optical magnetization reversal taking place on the femtosecond time scale. Therefore, it implies an ultrafast transfer of angular momentum from the spin system to another degree of freedom such as the orbital momentum of electrons or lattice. In turn this also implies a strong spin-orbit interaction during the optical excitation. Such fast coupling is in agreement with the recent experimental demonstration of a laser enhanced spin-orbit coupling in Ni.⁴ Hence we believe, as also discussed in Ref. 2, that relaxation of the non-equilibrium d electrons plays an important role in increasing the number of channels available to relax the magnetization through spin-orbit coupling and momentum relaxation.⁷ A subpicosecond switching rate also indicates that relaxation effects induced by optical excitations are much larger than those derived from FMR measurements. This is in line with the different energy scales of 1 eV and 1 meV for optical switching and FMR, respectively.
- (2) How is it possible that the angular momentum of the photons can efficiently change magnetization? The opti-

cal electric dipole transition cannot affect the electronic spin. Magnetic dipole transition may affect the spin but it requires annihilation of the photon. However, there are not enough photons in the laser pulse to provide enough angular momentum for a magnetization reversal.⁸ An efficient switching mechanism may take place via a stimulated Raman-like scattering process.⁵ Yet, in order to be effective, this switching mechanism requires heating of the spin system to temperatures close to Curie temperature.¹ As we will further demonstrate, the all-optical switching takes place even at a temperature of 200 K lower than that used in Ref. 1. Based on this result, a more realistic scenario may be accounted to an optical Barnett effect which works best at zero temperature.²

- (3) Another issue that needs to be discussed is how the femtosecond helicity-induced coherence among the itinerant electrons may be converted to a static magnetization. It is well known that the lifetime of a state at 1 eV above the Fermi level is a few femtoseconds.⁹ Relaxation of these excited states leads to the destruction of the spin coherence via inelastic scattering processes. Indeed, it is by now well accepted that there is no helicity-induced magnetization reversal in pure transition-metals such as Ni, under similar conditions.¹⁰ Thus, the observed memory effect in our experiments may be explained as follows. During the optical excitation, the negative exchange between the excited d electrons of both Gd and Fe collapses.¹¹ On the other hand, the rare-earth $4f$ bands are about 4 eV below the Fermi level and are therefore not directly excited by the 1.5 eV photon energy used in our experiments. Yet, a strong hybridization between the d -spins and the f -spins of the rare earth causes the component of the f -spins along the chirality of the laser to change sign at high enough laser powers. Therefore, although the spin coherence of the excited states may be lost via relaxation in electron-electron scattering, f -spins will maintain sufficient coherence in the reversal process and therefore maintain information about the chirality of the laser. In this way, f -spins will serve as nucleation points for what will later become, via processes such as domain wall propagation, a complete reversed magnetic domain. This explains the importance of the rare earth moments. A convincing test of this model will be the investigation of the all-optical switching in SmCo_5 and YCo_5 . The former has uncompensated f -spins but the latter does not while both have similar crystal structure and magnetocrystalline anisotropy.

Next we discuss the low temperature measurements. The same sample has been placed into a cryostat and exposed to 40 fs circularly polarized laser pulses generated at a repetition rate of 1 kHz. The laser wavelength was 800 nm and the laser helicity has been chosen to be that relevant for the switching. Initially at 300 K, in order to indicate the location of the laser spot onto the sample, GdFeCo has been exposed to a laser fluence of about 3 mJ/cm² for a few seconds. Next, the laser fluence has been reduced to about 2.5 mJ/cm², that is below the fluence threshold required for the all-optical switching in this sample.¹ At this laser fluence

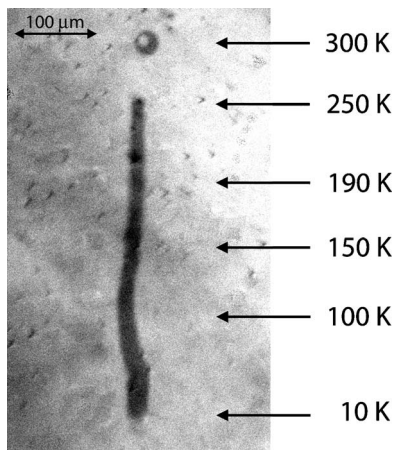


FIG. 2. All-optical switching of magnetization as a function of temperature, in GdFeCo. Black and white areas correspond to oppositely oriented magnetic domains, perpendicular to the sample plain. The dotlike magnetic domain was created in order to indicated the laser beam location onto the sample, at 300 K. The switching behavior as a function of temperature has been investigated at a laser fluence of 2.5 mJ/cm^2 . For this laser fluence no switching is observed at room temperature but it becomes visible only when the temperature is reduced below 250 K.

no switching could be observed. Under these conditions the temperature on the sample was reduced down to 10 K at a rate of about 5 K/min while the sample was slowly vertically shifted (from down to up). One can observe in Fig. 2 that while from 300 down to 250 K the laser fluence of 2.5 mJ/cm^2 does not change the magnetic state of the sample, the conditions change drastically below 250 K. More specifically, below this temperature the switching spot appears and increasingly broadens as the temperature is reduced down to 10 K. This observation gives clear evidence that thermal fluctuations introduce decoherence in the system that upsets the reversal process. Hence, increasing the temperature decreases the efficiency of the all-optical switching which appears to invalidate the original ideas based on stimulated Raman-like scattering.

In conclusion, we have demonstrated that all-optical switching in GdFeCo is not a thermally assisted process but in fact thermal fluctuations degrade the efficiency of the reversal. Moreover, we showed that the switching occurs on a subpicosecond time scale which cannot be explained in terms of the much slower precessional reversal. Both findings, i.e., fast and athermal switching mechanism, make all-optical recording on rare-earth transition metal alloys a promising technique for magnetic storage with high data rates. Furthermore, the all-optical switching using the long 500 fs laser pulses shown here, together with the recent demonstration of miniature plasmonic wave plates,¹² push the all-optical switching close to applications.

We would like to thank Professor A. Tsukamoto for preparing the samples and Dr. Paul Jones for his assistance with the laser set up. C.D.S. expresses his gratitude to Professor Rasing for mentorship. A.R. acknowledges fruitful discussions with Professor Chantrell.

¹C. D. Stanciu, F. Hansteen, A. V. Kimel, A. Kirilyuk, A. Tsukamoto, A. Itoh, and Th. Rasing, *Phys. Rev. Lett.* **99**, 047601 (2007).

²A. Rebei and J. Hohlfeld, *Phys. Lett. A* **372**, 1915 (2008); *J. Appl. Phys.* **103**, 07B118 (2008).

³S. J. Barnett, *Phys. Rev.* **6**, 239 (1915).

⁴C. Stamm, T. Kachel, N. Pontius, R. Mitzner, T. Quast, K. Holldack, S. Khan, C. Lupulescu, E. F. Aziz, M. Wietstruk, H. A. Dürr, and W. Eberhardt, *Nature Mater.* **6**, 740 (2007).

⁵C. D. Stanciu, Ph.D. thesis, Radboud University Nijmegen, 2008.

⁶J. Hohlfeld, Th. Gerrits, M. Bilderbeek, and Th. Rasing, *Phys. Rev. B* **65**, 012413 (2001).

⁷A. Rebei and J. Hohlfeld, *Phys. Rev. Lett.* **97**, 117601 (2006).

⁸B. Koopmans, M. van Kampen, J. T. Kohlhepp, and W. J. M. de Jonge, *Phys. Rev. Lett.* **85**, 844 (2000).

⁹M. Aeschlimann, M. Bauer, S. Pawlik, W. Weber, R. Burgermeister, D. Oberli, and H. C. Siegmann, *Phys. Rev. Lett.* **79**, 5158 (1997).

¹⁰F. Dalla Longa, J. T. Kohlhepp, W. J. M. de Jonge, and B. Koopmans, *Phys. Rev. B* **75**, 224431 (2007).

¹¹H.-S. Rhie, H. A. Dürr, and W. Eberhardt, *Phys. Rev. Lett.* **90**, 247201 (2003).

¹²A. Drezet, C. Genet, and T. W. Ebbesen, *Phys. Rev. Lett.* **101**, 043902 (2008).