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Single-pulse magneto-optic microscopy: a new tool for studying optically induced magnetization reversals

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We have developed a femtosecond magneto-optical imaging system that allows measurements of permanent magnetic effects that are initiated by a single excitation pulse. The system combines a subpicosecond temporal resolution and a high spatial resolution. We demonstrate the system in an experiment that studies the laser-induced magnetization reversal in ferromagnetic thin films. © 2008 Optical Society of America

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The desire to improve magnetic recording devices and to make them more efficient has motivated many recent explorations of the magnetization dynamics initiated by short-pulse laser excitation. Many studies have been reported on itinerant ferromagnets such as nickel, Permalloy, and cobalt alloys, and most of these focused on the demagnetization process that occurs on femtosecond and picosecond time scales [1–6]. In these experiments the local magnetization was probed, and the dynamics of magnetic reversal, its spatial extent, and the effects of magnetic domains were ignored. Other experiments studied the laser-induced reversal, using magneto-optical imaging techniques that lacked a time-resolved capability [7]. Recently this shortcoming was addressed with a high-resolution scanning femtosecond microscope, designed specifically for studying individual quantum dots [8]. For that purpose, however, the excitation and probe pulses were scanned together, and the measurement remained essentially local.

From the above it is clear that more extensive studies of the magnetization reversal in response to short-pulse laser excitation are essential. In particular, imaging techniques that are sensitive to the magnetic contrast, such as magneto-optical Kerr microscopy, should be combined with femtosecond laser based imaging systems [9] to allow detailed studies of the magnetization reversal process in response to short-pulse laser excitation. Experimental limitations make these measurements quite challenging, though. The reason for this is the fact that the reversal appears to occur in response to a single pump pulse, and is, in this sense, “irreversible,” while most experimental techniques rely on averaging over hundreds and even thousands of pulses to achieve an acceptable signal to noise ratio. A few researchers have addressed the irreversibility problem [10], but their experimental techniques lacked the spatial dimension element.

Here we describe an innovative magneto-optical imaging system (Fig. 1) that we have developed, which combines single-pulse measurements with 100 fs temporal resolution with a spatial resolution up to ≈50 μm, and thus allows us to explore the reversal dynamics in detail. We use a 70 fs, 1 kHz Ti:sapphire regenerative amplifier and a slightly modified time-resolved pump–probe setup, in which the probe spot size cover a large area of the sample, which extends far beyond the area excited by the pump beam. The probe pulse therefore acts as a 70 fs illumination source (or “flash”). The magnetization measurements rely on the magneto-optical Kerr effect (MOKE), using conventional optical polarizers. The sample is placed within the coils of an electromagnet, and the magnetic field is varied using a computer-controlled power supply that adjusts the current through the electromagnet. A single lens, with a focal length of 75 mm, is used to image the sample onto a Si CCD camera with 1024 × 128 pixels. Each pixel is 26 μm × 26 μm, and the optical magnification is 10×. To eliminate scattering from the pump beam into the CCD an iris may be placed behind the imaging lens, with a corresponding reduction of the numerical aperture and a loss of spatial resolution.

Each image acquired by the CCD camera represents the spatial distribution of the transmitted probe. To retrieve the spatial magnetization distribution, a reference image corresponding to a negative field is subtracted from the data. The images are

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Fig. 1. (Color online) Time-resolved MOKE microscope described in the text. “1” and “2” denote the alternative shutter positions for fast (pump–probe) and slow (time exposure) measurements, respectively.
then normalized by the full contrast image, obtained by subtracting an image taken at maximum negative field from an image taken at maximum positive field, to account for the uneven illumination of the probe beam. The residual pump scattering is removed by subtracting a reference image obtained by exposing the camera to the same number of pump pulses (with the probe beam blocked).

The above optical setup is combined with a unique electronic system specifically designed for single-pulse measurements. It includes a current source for the magnet, an electromechanical shutter, and an electronic card that synchronizes them. The electronic system drives the current through the magnet so as to reset the magnetization of the sample before each exposure and then opens the shutter to allow any given number of pulses in each exposure. This process is typically repeated a few hundred times while the CCD remains active to improve the signal to noise ratio. Thus we can measure the effect of any number of pulses down to a single pulse. Furthermore, measurements can be carried out in time delay regimes that cover many orders of magnitude: the femtosecond and picosecond regimes are explored using standard mechanical delay lines, while measurements in the millisecond regime are performed by setting a negative delay (i.e., the probe pulse precedes the pump pulse), and subtracting images acquired with single pulse pair excitations from images obtained by exposing the sample to pairs of pump and probe pulses. Measurements at very long delays (e.g., seconds) are possible by electronic timing of the camera exposure. Measurements in the microsecond regime are also possible, using a different, electronically triggered probe laser (such measurements are out of the scope of this Letter, however).

The magneto-optical imaging system described above allows us to address fundamental questions about the physics behind the magnetization reversal that is initiated by a single laser pulse. For example, it is unclear if the reversal is localized around the laser-excited spot, or if it acts as a nucleation center, from which the reversal may propagate. It is also unclear if a single pulse is indeed always sufficient to reverse the magnetization, and how this may depend on the applied field and the laser fluence (one cannot assume that the temporal dynamics that were observed in GdFeCo [10] are general properties of ferromagnets as well as ferrimagnets). The effects of successive pulses are also unclear, as are the consequences of the relative sizes of the laser spot and the typical magnetic domain. The longtime stability of the reversal, as a function of the applied field, is also unknown. In this Letter we focus on the spatiotemporal dynamics of the laser-induced magnetic reversal in Permalloy thin films. The results described below were obtained at room temperature with a 20-nm-thick film of Ni$_{0.8}$Fe$_{0.2}$ deposited on a GaAs substrate. In equilibrium the magnetization lies in the plane of this sample. The sample is first magnetized in one direction (“negative field”), and the effect of the pump pulse is then measured at a set reverse field (i.e. “positive”) below the coercive field.

Before presenting the response of the sample to the optical excitation we show the pattern of slow spontaneous magnetization reversal as a function of time at an external magnetic field of $H_0 \approx 1.2$ Oe. The different images are shown in Fig. 2. As the figure shows, on the time scale of the measurements there is no spontaneous reversal at the center of the image. We therefore aimed the pump beam at the center of the image and investigated the optically induced magnetization reversal at a field of $H_0 \approx 1.2$ Oe. For comparison, we have also measured the response at $H=0$. The results obtained for a pump fluence of 48 mJ/cm$^2$, as a function of the time delay, are shown in Fig. 3. Figure 3(a) shows that the magneto-optical signals at zero field and at $H_0$ are indistinguishable in the picosecond regime, but a permanent reversal is obtained (with this fluence) in the millisecond regime, only at $H_0$. Figure 3(b), also obtained with a fluence of 48 mJ/cm$^2$ and at $H_0$, shows a slow spatial expansion of the reversal as a function of time, along with spontaneous reversal on the right-hand side of the image. The data show that the picosecond regime...

![Fig. 2](image-url)  
Fig. 2. Slow spontaneous magnetization reversal in a Permalloy sample at a field of $H_0 \approx 1.2$ Oe. The elapsed times from the moment the field was set are indicated.

![Fig. 3](image-url)  
Fig. 3. (a) Pump–probe measurements at different time delays, at zero field and at $H_0$, with a pump fluence of 48 mJ/cm$^2$. Permanent reversal is obtained only at $H_0$ (in these measurements an ~0.2 cm diameter iris was used, yielding a resolution of ~50 μm); (b) slow measurements carried out in the timed exposure configuration. The elapsed times from the moment of excitation by a single pump pulse are indicated. Spontaneous reversal can be seen on the right of the images.
is dominated by thermal effects and that the actual reversal occurs over much longer times compared to the reversal time observed in experiments on ferromagnets [10].

Based on the known equilibrium specific heat of Permalloy, we conclude that the fluence of 48 mJ/cm² corresponds to a temperature rise of \( \approx 700^\circ K \). No permanent reversal was observed in our experiments at lower pump fluences. This implies that the sample must reach the Curie temperature for a permanent reversal to occur, even at fields that approach the coercive field. The detailed behavior as a function of field and fluence is, however, beyond the scope of this Letter and will be discussed elsewhere.

The data presented above showed the local effect of the optical excitation. In the following we demonstrate that the optically induced reversal also has a nonlocal effect on the spontaneous propagation of domain walls at later times. Figure 4 shows two examples of the nonlocal effects of a single pump pulse. In Fig. 4(b) the excitation spot is inside a “soft” region, i.e., the spontaneously reversed area seen in Fig. 4(a). A comparison of panels (a) and (b) shows that the pump-induced reversal acts as a nucleation center, from which the reversal propagates outwards, to fill the same portion of the sample that would otherwise reverse spontaneously. This process reaches a steady state after \( \approx 3 \) min, compared to \( \approx 6 \) min without excitation. A comparison of Fig. 4(c) to Fig. 2 shows the effect of excitation in a “hard” portion of the sample next to the soft region. In this case the optical excitation expedites the propagation of nearby domain walls, while slowing the propagation in other parts of the image. These measurements show that the pump excitation modifies the energy landscape in its vicinity. Importantly, there are areas in which the pump-induced reversal lowers the energy barrier, while in other areas the energy barrier is increased.

In conclusion, we have developed a new experimental tool that allows us to investigate magnetization dynamics in magnetic thin films with high temporal and spatial resolutions. We have used this tool to explore the optically induced magnetization reversal in Permalloy and studied its effect on magnetic domain processes. The dependence of the observed phenomena on excitation parameters such as the field and the laser fluence is under investigation.

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