Helicity and field dependent magnetization dynamics of ferromagnetic Co/Pt multilayers


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Microscopic model for all optical switching in ferromagnets
The discovery of all-optical switching (AOS) in amorphous GdFeCo alloys\(^1\) raised many questions about the nature as well as the generality of this phenomenon. Attempts of engineering new materials suitable for both AOS and technological applications\(^2\) resulted in the surprising observation of a helicity-dependent all-optical magnetization reversal\(^3\) in a number of ferromagnetic multilayers with a perpendicular magnetic anisotropy.\(^4,5\) This effect was particularly found in Co/X (\(=\) Pt, Pd, \(\ldots\)) multilayers, a class of materials known for their strong thermal stability\(^6\) and their potential for patterned magnetic media\(^7,8\) or spin-transfer torque based memory\(^9,10\) applications. The enhanced spin-orbit coupling at the Co/Pt or Co/Pd interfaces was shown to cause an increased rate of laser-induced demagnetization\(^18\) compared to that of pure transition metal thin films.\(^11,12\) Further, a dependence of the demagnetization rate on the thickness of the transition metal in the multilayers was detected.\(^19\)

The laser-induced magnetization reversal observed in these ferromagnetic materials\(^1\) does not fit the paradigm of the existing understanding of AOS in ferrimagnets.\(^20–23\) The data reported in Ref. \(^3\) only revealed the final states of the ferromagnetic samples after being excited by multiple laser pulses. So far, neither the actual mechanism nor the timescale of this reversal is known. Moreover, a very recent study of Co/Pt multilayers revealed a cumulative nature of the magnetization reversal with a “certain number of laser pulses needed” to obtain a full and reproducible helicity-dependent AOS.\(^24\) In that work, the authors employed electrical characterization of the AOS in Co/Pt structures using a Hall bar, but this approach neither gave insight in the effect of a single pulse nor into its dynamics or mechanism.

To understand what happens to the magnetization after a single laser pulse excitation, we studied the magnetization dynamics of ferromagnetic Co/Pt multilayers. The structures demonstrated the same helicity-dependent magnetization reversal as reported in Ref. \(^3\). Employing single-shot time-resolved magneto-optical Kerr effect imaging, our study detects no signatures of AOS after a single 60 fs optical laser pulse excitation, but reveals an ultra-fast demagnetization followed by a magnetization recovery. Full demagnetization occurs within 1 ps after laser excitation. The magnetization dynamics reveals a small helicity dependence caused by magnetic circular dichroism. When an external magnetic field is applied, a heat-assisted magnetization reversal occurs on a nanosecond time scale.

The [Co(0.4 nm)/Pt(0.7 nm)]\(^3\) multilayers were fabricated by DC ultra-high vacuum magnetron sputtering using a conical sputter-up geometry from an AJA International ATC-2200 system with the targets tilted and arranged in a circle around a central target (here Pt).\(^25\) The substrate, a (100) oriented Si 1-in. wafer with a native SiO\(_2\) surface layer, rotates during deposition at \(f_{\text{sub}} \approx 3\) Hz and is at the focal point of the targets. We used a Ta(1.5 nm) adhesion layer on top of the native SiO\(_2\) surface layer and a Pt(20.0 nm) seed layer to obtain a (111) texture with a mosaic spread of \(\delta \approx 5^\circ\) full width at half maximum for the multilayer out-of-plane Bragg reflection. The samples were taken out of the chamber after the deposition of a Pt(2 nm) cap layer. The multilayers were deposited at 3 mTorr and 8.5 mTorr of argon pressure to slightly tune the magnetic switching behavior. Changes of the deposition pressure allows tuning the film microstructure from a continuous film to magnetically isolated grains.\(^28\) All the samples demonstrated perpendicular magnetic anisotropy and 100% remanence. The magneto-optical static characterization revealed an increase of the coercive field from \(H_c = 30\) mT to \(H_c = 110\) mT.
with increasing argon pressure from 3 mTorr to 8.5 mTorr. In the magnetization dynamics study, we could not detect any substantial difference between these various samples.

For the magnetization dynamics study, a single-shot time-resolved magneto-optical imaging setup, similar to the one used in Refs. 29 and 30, was employed. During the experiment, the multilayer was excited by a single circularly polarized 60-fs laser pulse with a central wavelength of \( \lambda = 800 \text{ nm} \), a beam-radius of \( \sigma \approx 80 \text{ \mu m} \), and a fluence ranging from 1 mJ/cm\(^2\) to 14 mJ/cm\(^2\). The magnetization state was probed by a time-delayed single linearly polarized laser pulse \( (\tau \approx 60 \text{ fs}, \lambda = 400 \text{ nm}, \sigma \approx 1 \text{ mm}, \text{ and a laser fluence} < 0.1 \text{ mJ/cm}^2) \). Employing the magneto-optical Kerr effect, domains with the magnetization parallel or antiparallel to the sample normal are seen as bright or dark regions, respectively, in the images recorded by a CCD camera. Next, we restored the initial conditions by applying an external magnetic field \( (H_{ext}) \) perpendicularly to the sample surface. Then, we switched the external magnetic field off for the next excitation/reading event \( (H_{ext} = 0 \text{ mT}) \). Alternatively, we can apply a constant magnetic field, smaller than the coercive field \( (H_{co} < H_c) \) during the whole excitation/reading process, to study the heat-assisted magnetic field-induced magnetization reversal in the structure. All the experiments were conducted at room temperature.

Fig. 1(a) demonstrates a sequence of selected magneto-optical images, acquired after illuminating the multilayer by a right circularly polarized (RCP) laser pulse. It represents the magnetic state before the optical excitation, as well as at \( \tau_{pp} = 60 \text{ ps}, 1735 \text{ ps}, \text{ and } 3300 \text{ ps after it. The scale-bar corresponds to } 70 \text{ \mu m}. A shrinking of the pump-induced spot is observed from the sequence of images. To get a measure of the photo-induced magnetization \( M_0 \) from each image, we averaged the pixels color value within a fixed region of the laser-induced area. We selected a region that corresponded to \( 45 \times 45 \text{ \mu m}^2 \) and used it for all the images obtained during the experiment. To be able to calibrate the \( M_0 \) signal, before any laser excitation we applied an external magnetic field, saturating the sample magnetization parallel to the sample normal. Then, the external magnetic field was switched off, and we averaged the pixels color value ascribed to the \( M_0 \) state. By repeating the procedure for the opposite direction of the magnetic field, we determined the \( M_{z,\perp} \) signal. We defined an initial magnetization value before the laser excitation as \( M_0 = (M_{z,\parallel} - M_{z,\perp})/2 \). We normalized the photo-induced magnetization \( M_z \) for every image taken at various time delays between pump and probe laser pulses, and reconstructed the magnetization dynamics by plotting the \( M_z(t)/M_0 \) value as a function of time. Figs. 1(b) and 2 demonstrate the details of the magnetization dynamics of the Co/Pt multilayer studied as a function of the external magnetic field (Fig. 1(b)) and laser pump fluence (Fig. 2). All data reveal an ultra-fast pump-induced quenching of the magnetization, typical for ferromagnetic materials, followed by a magnetization relaxation in the absence of an external magnetic field. In the lower laser fluence regime, the magnetization is able to relax completely to the initial state within at least \( T_\tau \approx 500 \text{ ps after the laser pulse excitation} \) (see inset in Fig. 1(b)). In the high laser fluence regime (higher than 10 mJ/cm\(^2\)), a fully demagnetized magnetic system does not recover to the initial state without an external magnetic field applied. After the relaxation, the averaged final state is always somewhat demagnetized, which can be explained by the creation of a multi-domain state by the laser excitation, with magnetic domains oriented randomly within the

FIG. 1. Reconstructed magnetization dynamics of \([\text{Co(0.4 nm)/Pt(0.7 nm)}]_3\) ferromagnetic multilayer, triggered by a single 60 fs circularly polarized laser pulse excitation with a fluence of 12 mJ/cm\(^2\). (a) A sequence of selected magneto-optical images, obtained at different delays between pump and probe without an external magnetic field applied. The scale bar is 70 \text{ \mu m}. (b) The magnetization dynamics extracted from the obtained images on long and short (see inset) time-scales. With no external magnetic field applied \( (H_{ext,0}) \), a full ultrafast demagnetization occurs within 1 ps and is followed by a slow magnetization recovery. In case a smaller than a coercive field is applied perpendicular to the sample surface \( (|H_{ext}| = |H_{co}| = 10 \text{ mT}) \), the heat-assisted magnetization reversal is detected.

FIG. 2. The laser fluence dependence of the magnetization dynamics reconstructed from magneto-optical images (see calibration procedure explanation in the text). The ultrafast demagnetization scales with the laser fluence.
excitation area. Due to the spatial resolution limit, this random distribution is seen by the CCD camera as an averaged single color state, which corresponds to a partial demagnetization.

By applying an external magnetic field $H_{ext} = 10 \text{ mT}$ in these multilayers (Fig. 1(b)). The laser pulse heats the system up changing the intrinsic material magnetic properties, such as magnetization saturation, magnetic anisotropy, and coercivity. With the coercivity lowered, the external magnetic field is capable to steer the magnetization of the heated area towards the direction parallel to the field. For the excitation fluence of 12 mJ/cm$^2$ and the external magnetic field of $H_{ext} = 10 \text{ mT}$ in opposite direction, around 60% of the full magnetization reversal was observed at the sample normal. By pumping the sample by a RCP pulse, a state compared to the initial state, but with a higher recovery rate than in the case of no magnetic field applied.

To address the possible helicity dependence of the demagnetization dynamics, we illuminated the sample by a 60 fs left-(LCP) or right-circularly polarized (RCP) single laser pulse in the absence of an external magnetic field. Before the laser excitation, the magnetization of the sample was saturated either parallel ($\vec{M}_1$) or antiparallel ($\vec{M}_{\perp}$) to the sample normal. By pumping the sample by a RCP pulse, a higher demagnetization efficiency is seen of the $\vec{M}_1$ initial state compared to the $\vec{M}_{\perp}$ state. Then, we excited the system by a LCP laser pulse which resulted in a more efficient demagnetization of the $\vec{M}_1$ initial state compared to the $\vec{M}_{\perp}$ one, which is represented in Fig. 3. This finding can be ascribed to magnetic circular dichroism. A distinct RCP and LCP laser pulse absorption leads to a different amount of heat transferred from the laser excitation to the system, which, consequently, results in an effective increase of the laser fluence for one of the polarizations and their different demagnetization efficiencies. This laser helicity-dependence on the magnetization dynamics in Co/Pt multilayers is similar to the one detected by Vahalpr et al. in GdFeCo amorphous alloys, which explained the narrow switchability window for those materials.

In conclusion, in this work we studied the single-shot helicity and field dependent magnetization dynamics of ferromagnetic [Co(0.4 nm)/Pt(0.7 nm)]$_3$ multilayers which have a perpendicular magnetic anisotropy and are suitable for multiple-pulse helicity-dependent magnetization reversal as reported in Ref. 3. Exciting the system by a single 60 fs circularly polarized laser pulse, an ultrafast demagnetization followed by a magnetization recovery was observed. The full ultrafast demagnetization occurred within 1 ps after laser pulse excitation of 12 mJ/cm$^2$. After a single circularly polarized optical pump excitation, a full magnetization recovery was seen in the lower (<9.07 mJ/cm$^2$) fluence regime. Exciting the sample by laser pulses with different polarization helicities, we detected a difference in the demagnetization efficiency for different initial magnetic states. The RCP single laser pulse excitation turned out to demagnetize the $\vec{M}_1$ state better than the $\vec{M}_{\perp}$ state, while for the LCP excitation the opposite was seen.

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