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

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We present helicity and field dependent magnetization dynamics of ferromagnetic Co/Pt multilayers, suitable for all-optical helicity-dependent switching. Employing single-shot time-resolved magneto-optical Kerr effect imaging, our study demonstrates an ultra-fast quenching of the magnetization after a single 60 fs laser pulse excitation followed by a 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 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[4–6]. This effect was particularly found in Co/X (=Pt, Pd, ...) multilayers, a class of materials, known for their strong thermal stability[7], and their potential for patterned magnetic media[8,9] or spin-transfer torque based memory[10–13] 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[14–17]. 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[3] does not fit to 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. Neither the actual mechanism nor the timescale of this reversal is known so far. 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 did neither give 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 a single 60 fs laser pulse excitation of sufficient fluence. Most importantly, unlike in previous studies of ferromagnets[25,26], we were able to detect an, albeit small, helicity dependence of the magnetization dynamics after a single laser pulse excitation. This dependence can be explained by the magnetic circular dichroism (MCD) of the material. Additionally, our study demonstrates that heat-assisted magnetization reversal occurs after a single femtosecond laser pulse excitation on a nanosecond time scale when an external magnetic field is applied.

The $[Co(0.4nm)/Pt(0.7nm)]_3$ multilayers were fabricated by DC ultra-high vacuum magnetron sputtering using a confocal 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)[27]. The substrate, a (100) oriented Si 1-inch wafer with a native SiO_2 surface layer, rotates during deposition at $f_{sub} \approx 3$ Hz and is at the focal point of the targets. We used a Ta(1.5nm) adhesion layer on top of the native SiO_2 surface layer and a Pt(20.0nm) seed layer to obtain a (111) texture with a mosaic spread of $\alpha \approx 5°$ 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(2nm) 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 be-

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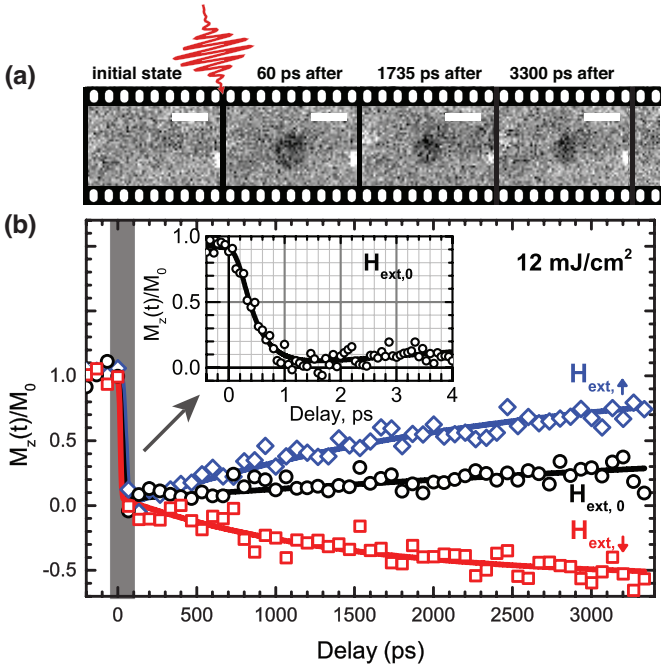

FIG. 1. (Color Online) Reconstructed magnetization dynamics of $[Co(0.4nm)/Pt(0.7nm)]_3$ ferromagnetic multilayer, triggered by a single 60 fs circularly polarized laser pulse excitation with a fluence of 12 mJ/cm^2 (a) A sequence of selected magneto-optical images, obtained at different delays between pump and probe without an external magnetic field applied. The scale bar is 70 μm. (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 perpendicularly to the sample surface ($|\mathbf{H}_{ext,\uparrow}| = |\mathbf{H}_{ext,\downarrow}| = 10mT$), the heat-assisted magnetization reversal is detected.

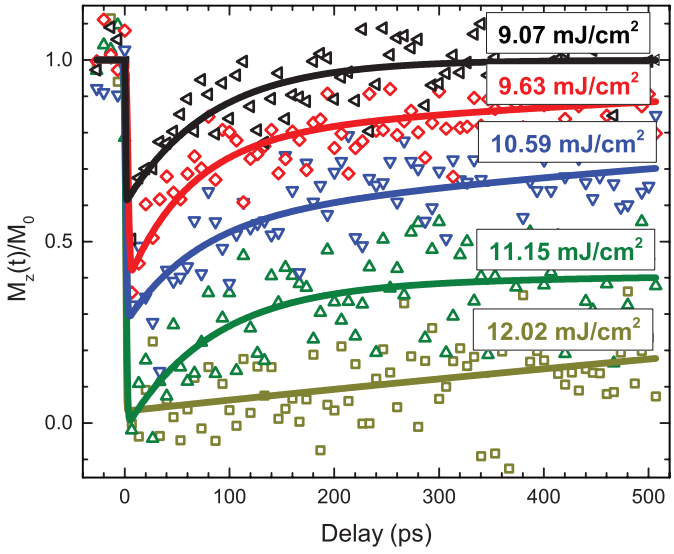

FIG. 2. (Color Online) The laser fluence dependence of the magnetization dynamics reconstructed from magneto-optical images (see calibration procedure explanation in the text). The ultrafast demagnetization scales with the laser fluence.

tween these various samples.

For the magnetization dynamics study, a single-shot time-resolved magneto-optical imaging setup, similar to the one used in Ref. 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 $\lambda = 800$ nm, a beam-radius of $\sigma \approx 80$ μm, 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 ($\tau \approx 60$ fs, $\lambda = 400$ nm, $\sigma \approx 1$ mm, and a laser fluence < 0.1 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 ($H_{ext} = 0$ mT). Alternatively, we can apply a constant magnetic field, smaller than the coercive field ($H_{ext} < H_c$) 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 laser pulse. It represents the magnetic state before the optical excitation, as well as at $\tau_{pp} = 60$ ps, 1735 ps, and 3300 ps after it. The scale-bar corresponds to 70 μm. A shrinking of the pump-induced spot is observed from the sequence of images. To get a measure of the photo-induced magnetization M_z 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 $45\times45\mu m^2$, and used it for all the images obtained during the experiment. To be able to calibrate the M_z 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 $M_{z,\uparrow}$ state. By repeating the procedure for the opposite direction of the magnetic field, we determined the $M_{z,\downarrow}$ signal. We defined an initial magnetization value before the laser excitation as $M_0 = (M_{z,\uparrow} - M_{z,\downarrow})/2$. 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. Fig. 1 (b) and Fig. 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[16], fol-

lowed 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 $\tau_r \approx 500$ ps after the laser pulse excitation (9.07 mJ/cm^2 in Fig. 2). The ultrafast demagnetization efficiency is seen to be proportional to the pump fluence, with full demagnetization occurring within $\tau_0 \approx 1$ *ps* for a fluence of 12 mJ/cm^2 (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 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 $H_{ext} = 10$ mT $< H_c$ we were able to demonstrate heat-assisted magnetization reversal[31] 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 $H_{ext} = 10$ mT in opposite direction, around 60% of the full magnetization reversal was observed at $\tau_{pp} = 3.3$ ns after a laser pulse excitation. Therefore, these results demonstrate, that the $[Co(0.4nm)/Pt(0.7nm)]_3$ structure is suitable for heat-assisted magnetization reversal. Note that a similar heat-assisted magnetization reversal was observed in Co/Pd multilayers[32], materials used in Hard Disk Drive industry in the early days. With a magnetic field of H_{ext} =10 mT applied in the original direction, the ultrafast demagnetization is followed by a magnetization relaxation towards the initial state, but with a higher recovery rate than in case of no magnetic field applied.

To address the possible helicity dependence of the (de)magnetization 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 ($\vec{M}_\uparrow$) or antiparallel ($\vec{M}_\downarrow$) to the sample normal. By pumping the sample by a RCP pulse, a higher demagnetization efficiency is seen of the $\vec{M}_\downarrow$ initial state compared to the $\vec{M}_\uparrow$ state. Then we excited the system by a LCP laser pulse which resulted in a more efficient demagnetization of the $\vec{M}_\uparrow$ initial state compared to the $\vec{M}_\downarrow$ 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

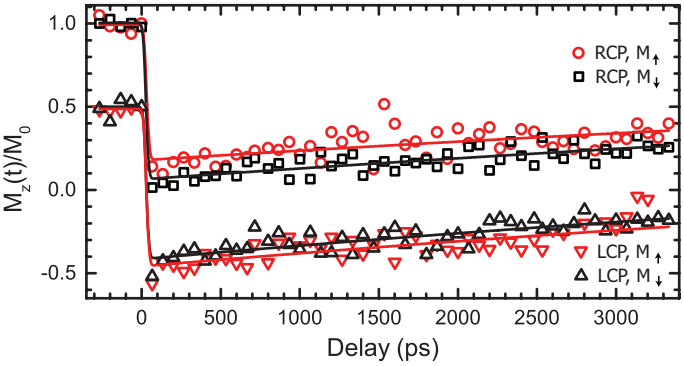

FIG. 3. (Color Online) Polarization dependence of the magnetization dynamics after a single 60 fs laser pulse excitation. The RCP and LCP optical pump pulses are used to excite the multilayer, which magnetization is saturated parallel $\vec{M}_\uparrow$ or antiparallel $\vec{M}_\downarrow$ to the sample normal. The magnetization dynamics for LCP laser excitation is shifted down by 0.5 for a better visual representation.

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 Vahaplar et. al in GdFeCo amorphous alloys[30], 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 $[Co(0.4nm)/Pt(0.7nm)]_3$ 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 12mJ/cm^2. After a single circularly polarized optical pump excitation, a full magnetization recovery was seen in the lower (< 9.07mJ/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 $\vec{M}_\downarrow$ state better than the $\vec{M}_\uparrow$ state, while for the LCP excitation the opposite was seen. We explain this observation by magnetic circular dichroism of this multilayer structure. Applying an external magnetic field smaller than the coercive field ($H_{ext} = 10$ mT $< H_c$), the magnetization could be reversed after a single 60 fs laser pulse excitation. This heat-assisted magnetization reversal occurred on the time scale of nanoseconds.

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[1] C. D. Stanciu, F. Hansteen, A. V. Kimel, A. Kirilyuk, A. Tsukamoto, A. Itoh, and Th. Rasing. *Phys. Rev. Lett.*, 99(4):047601, 2007.

[2] S. Mangin, M. Gottwald, C-H. Lambert, D. Steil, V. Uhlĩr, L. Pang, M. Hehn, S. Alebrand, M. Cinchetti, G. Malinowski, Y. Fainman, M. Aeschlimann, and E.E. Fullerton. *Nature Mat.*, 13(3):286292, 2014.

[3] C-H. Lambert, S. Mangin, B.S.D.Ch.S. Varaprasad, Y.K. Takahashi, M. Hehn, M. Cinchetti, G. Malinowski, K. Hono, Y. Fainman, M. Aeschlimann, and E.E. Fullerton. *Science*, 1253493, 2014.

[4] D. Weller, Y. Wu, J. Stohr, and M. G. Samant *Phys. Rev. B*, 49(18):12888, 1994.

[5] M. T. Johnson, P. J. H. Bloemen, F. J. A. den Broeder, and J. J. de Vries. *Rep. Prog. Phys.*, 59:1409, 1996.

[6] M. Gottwald, K. Lee, J. J. Kan, B. Ocker, J. Wrona, S. Tibus, J. Langer, S. H.Kang, and E. E. Fullerton *Appl. Phys. Lett.*, 102:052405, 2013.

[7] X. Chen, and M. H. Kryder *J. Appl. Phys.*, 85:5006, 1999

[8] T. Thomson, G. Hu, and B. D. Terris *Phys. Rev. Lett.*, 96:257204, 2006.

[9] O. Hellwig, A. Berger, T. Thomson, E. Dobisz, Z. Z. Bandic, H. Yang, D. S. Kercher, and E. E. Fullerton. *Appl. Phys. Lett.*, 90:162516, 2007.

[10] K.-F. Huang, D.-Sh. Wang, H.-H. Lin, and Ch.-H. Lai *Appl. Phys. Lett.* 107:232407, 2015

[11] S. Mangin, D. Ravelosona, J. A. Katine, M. J. Carey, B. D. Terris, and E. E. Fullerton. *Nature Mat.*, 5:210, 2006.

[12] A. D. Kent, and D. C. Worledge *Nature Nanotechnol.*, 10:187, 2015.

[13] S. Parkin, and S.-H. Yang *Nature Nanotechnol.*, 10:195, 2015

[14] G. P. Zhang, and W. Hbner *Phys. Rev. Lett.*, 85:3025, 2000.

[15] M. Cinchetti, M. Sanchez Albaneda, D. Hoffmann, T. Roth, J.-P. Wustenberg, M. Krau, O. Andreyev, H. C. Schneider, M. Bauer, and M. Aeschlimann *Phys. Rev. Lett.*, 97:177201, 2006.

[16] B. Koopmans, G. Malinowski, F. Dalla Longa, D. Steiauf, M. Fahnle, T. Roth, M. Cinchetti, and M. Aeschlimann. *Nature Mat.*, 9:259, 2010.

[17] T. Roth, A. J. Schellekens, S. Alebrand, O. Schmitt, D. Steil, B. Koopmans, M. Cinchetti, M. Aeschlimann *Phys. Rev. X*, 2:021006, 2012

[18] K. C. Kuiper, T. Roth, A. J. Schellekens, O. Schmitt, B. Koopmans, M. Cinchetti, and M. Aeschlimann *Appl. Phys. Lett.*, 105:202402, 2014.

[19] S. Pal, D. Polley, R. K. Mitra, and A. Barman. *Solid State Comm.*, 221:50, 2015.

[20] A. Kirilyuk, A. V. Kimel, and Th. Rasing. *Rep. Prog. Phys.*, 76(2):026501, 2013.

[21] J. H. Mentink, J. Hellsvik, D. V. Afanasiev, B. A. Ivanov, A. Kirilyuk, A. V. Kimel, O. Eriksson, M. I. Katsnelson, and Th. Rasing. *Phys. Rev. Lett.*, 108(5):057202, 2012.

[22] T. A. Ostler, J. Barker, R. F. L. Evans, R. W. Chantrell, U. Atxitia, O. Chubykalo-Fesenko, S. El Moussaoui, L. Le Guyader, E. Mengotti, L. J. Heyderman, F. Nolting, A. Tsukamoto, A. Itoh, D. Afanasiev, B. A. Ivanov, A. M. Kalashnikova, K. Vahaplar, J. Mentink, A. Kirilyuk, Th. Rasing, and A. V. Kimel *Nat. Comm.*, 3:666, 2012.

[23] I. Radu et al. *Nature*, 472(7342):205, 2011.

[24] M. S. El Hadri, P. Pirro, C.-H. Lambert, N. Bergeard, S. Petit-Watelot, M. Hehn, G. Malinowski, F. Montaigne, Y. Quessab, R. Medapalli, E. E. Fullerton, and S. Mangin *Appl. Phys. Lett.*, 108:092405, 2016

[25] F. Dalla Longa, J. T. Kohlhepp, W. J. M. de Jonge, and B. Koopmans *Phys. Rev. B*, 75:224431, 2007

[26] V. Lpez-Flores, J. Arabski, C. Stamm, V. Halt, N. Pontius, E. Beaurepaire, and C. Boeglin *Phys. Rev. B*, 86:014424, 2012

[27] O. Hellwig, T. Hauet, T. Thomson, E. Dobisz, J. D. Risner-Jamtgaard, D. Yaney, B. D. Terris, and E. E. Fullerton *Appl. Phys. Lett.*, 95:232505, 2009.

[28] M. S. Pierce, J. E. Davies, J. J. Turner, K. Chesnel, E. E. Fullerton, J. Nam, R. Hailstone, S. D. Kevan, J. B. Kortright, Kai Liu, L. B. Sorensen, B. R. York, and O. Hellwig. *Phys. Rev. B*, 87:184428, 2013.

[29] K. Vahaplar, A. M. Kalashnikova, A. V. Kimel, D. Hinzke, U. Nowak, R. Chantrell, A. Tsukamoto, A. Itoh, A. Kirilyuk, and Th. Rasing. *Phys. Rev. Lett.*, 103(11):117201, 2009.

[30] K. Vahaplar, A. M. Kalashnikova, A. V. Kimel, S. Gerlach, D. Hinzke, U. Nowak, R. Chantrell, A. Tsukamoto, A. Itoh, A. Kirilyuk, and Th. Rasing. *Phys. Rev. B*, 85(10):104402, 2012.

[31] M. H. Kryder, E. C. Gage, T. W. McDaniel, W. A. Challener, R. E. Rottmayer, G. Ju, Y-T. Hsia, and M. F. Erden. *Proc. of the IEEE*, 96(11):1810, 2008.

[32] A. Hirotsune, H. Nemoto, I. Takekuma, K. Nakamura, T. Ichihara, and B. Stipe. *IEEE Trans. on Magn.*, 46(6):1569, 2010.