Dear Editor,

We submit a manuscript entitled “Intriguing hysteresis dynamics in ultrafast photo-induced magnetization” to your journal, Physica Status Solidi b. Our main findings are

- ultrafast hysteresis behavior during a photo-induced demagnetization/remagnetization process for Co/Pt ferromagnetic multilayers.
- irreversible behavior in hysteresis at each stroboscopic measurement
- experimental demonstration that the stroboscopically measured hysteresis and the coercivity could be a stable parameter in describing the ultrafast photo-induced spin dynamics.

We believe that our work reports important experimental aspects in analyzing ferromagnetic materials on an ultrafast time scale.

Sincerely yours,
Dong-Eon Kim
Dong-Hyun Kim

Do you or any of your co-authors have a conflict of interest to declare? No. The authors declare no conflict of interest.
Abstract:
We have carried out time-resolved magneto-optical Kerr effect measurement to determine ultrafast hysteresis behavior during a photo-induced demagnetization/remagnetization process for Co/Pt ferromagnetic multilayers. Due to the stroboscopic measurement scheme, hysteresis curve measured by a pump-probe technique exhibits an irreversible behavior, repeatedly reset to a metastable hysteresis state at each stroboscopic measurement, observed as the reduction of the coercivity. We experimentally demonstrates that the stroboscopically measured hysteresis and the coercivity could be a stable parameter in describing the ultrafast photo-induced spin dynamics both for reversible and irreversible behavior. Moreover, we have found that an unusual behavior of coercivity behavior exists together with a nontrivial magnetization change under external fields comparable to the coercivity of the sample.
Intriguing hysteresis dynamics in ultrafast photo-induced magnetization

Je-Ho Shim¹, Chul-Hoon Kim³, Hong-Guang Piao⁴,⁵, Sang-Hyuk Lee⁴,⁶, Kyung Min Lee⁷, Jong-Ryul Jeong⁷, Seung-Young Park⁸, Yeon Suk Choi⁸, Dong Eon Kim¹,²a), and Dong-Hyun Kim²,⁴b)

¹Department of Physics and Center for Attosecond Science and Technology, POSTECH, Pohang 37673, South Korea
²Max Planck POSTECH/KOREA Research Initiative, Pohang, 37673, South Korea
³Department of Advanced Materials Chemistry, Korea University, Sejong 30019, South Korea
⁴Department of Physics, Chungbuk National University, Cheongju 28644, South Korea
⁵College of Science, China Three Gorges University, Yichang 443002, P. R. China
⁶Division of Industrial Metrology, Korea Research Institute of Standards and Science (KRISS), Daejeon 34113, South Korea
⁷Department of Material Science and Engineering and Graduate School of Energy Science and Technology, Chungnam National University, Daejeon 34134, South Korea
⁸Spin Engineering Physics Team, Korea Basic Science Institute, Daejeon 34133, South Korea

Abstract

We have carried out time-resolved magneto-optical Kerr effect measurement to determine ultrafast hysteresis behavior during a photo-induced demagnetization/remagnetization process for Co/Pt ferromagnetic multilayers. Due to the stroboscopic measurement scheme, hysteresis curve measured by a pump-probe technique exhibits an irreversible behavior, repeatedly reset to a metastable hysteresis state at each stroboscopic measurement, observed as the reduction of the coercivity. We experimentally demonstrates that the stroboscopically measured hysteresis and the coercivity could be a stable parameter in describing the ultrafast photo-induced spin dynamics both for reversible and irreversible behavior. Moreover, we have found that an unusual behavior of coercivity behavior exists together with a nontrivial magnetization change under external fields comparable to the coercivity of the sample.

a) kimd@postech.ac.kr
b) donghyun@cbnu.ac.kr
**Introduction**

Understanding of an ultrafast spin dynamics becomes essential for development of spintronic devices in operation with ultrafast speed as well as ultrahigh density [1-3]. Recently, photo-induced spin dynamics has been extensively investigated since Beurepaire et al. first reported a ultrafast demagnetization by femtosecond laser pulse in ferromagnetic Ni film by means of time-resolved magneto-optical Kerr effect (TR-MOKE) measurement [4]. Several mechanisms [5] such as electron-magnon interaction [6], Coulomb interaction [7], electron-phonon scattering [8], effect of magnetic domain [9], direct angular momentum transfer [10], hot-electron-induced demagnetization [11], and superdiffusive spin current [12] have been proposed to explain ultrafast demagnetization phenomenon. The exact underlying physics of the ultrafast spin dynamics is still under intense debates.

Generally, magnetic hysteresis curve is considered to be most important macroscopic feature in understanding ferromagnetic behavior of samples. For instance, coercivity, saturation field, remanence, and saturation magnetization provides basic characteristic parameters in analyzing overall ferromagnetic features. However, it has been known that magnetic hysteresis, measured during the photo-induced demagnetization/remagnetization process, is quite different from static hysteresis curves with much reduced coercivity [13-15]. The coercivity reduction is explained based on metastable states during the photo-induced demagnetization/remagnetization dynamics, leading to a so-called irreversible process, arising from the magnetization state change in metastable states [14,15]. To avoid the irreversible process issue in pump-probe stroboscopy measurement for ultrafast spin dynamics, external magnetic field cycling in synchronization with the same frequency as the stroboscopic repetition frequency was adopted in TR-MOKE setup [16,17]. With this synchronization, coercivity reduction becomes negligible since the metastable states at time-zero are replaced by the fully synchronized magnetic saturation.

On the other hand, interestingly, Shufa et al. have recently reported that coercivity measured by the pump-probe stroboscopy technique can be also reversible [15], where the field dependent Kerr signals are observed to be reversible near coercivity [16]. Thus, hysteresis behavior such as coercivity reduction in ultrafast spin dynamics still remains a scientific interest to further explore. Unfortunately, little has been known for detailed spin dynamics near the coercivity during the ultrafast photo-induced spin dynamics. Thus, rather simple magnetic system is required, where we can focus on the ultrafast photo-induced demagnetization...
behavior. Recently, ultrafast spin dynamics in multilayers with perpendicular magnetic anisotropy (PMA) have been explored [18-20], where the magnetization precession signal can be suppressed under external field normal to film plane so that only demagnetization and remagnetization is pronounced without extra precession signal. In this work, we have measured time-resolved metastable hysteresis curves during the photo-induced demagnetization/remagnetization process by means of pump-probe stroboscopic TR-MOKE measurement for ferromagnetic Co/Pt multilayer films, where both reversible and irreversible behavior are observed for Co/Pt multilayers with different repeat numbers. Unusual TR-MOKE signal under external fields comparable to the coercivity is observed even for the case of reversible process.

1. Experimental

We have measured TR-MOKE with variation of external fields as well as stroboscopic delay times. A CEP (carrier envelope phase)-stabilized multipass amplifier laser (Femtopower, Femtolaser. Inc.) was used as the light source for TR-MOKE. The pump and probe beam with 780 nm wavelength, 3 kHz repetition rate, and 25 fs pulse width are used. Both the pump and probe beams were set to have s-polarization. The effective time resolution is better than 200 fs, determined from the auto-correlation measurement. The details of the TR-MOKE set up are described elsewhere [21]. Magnetic hysteresis were measured at each delay time up to 700 ps. The pump laser fluence was kept to be 13.2 mJ/cm² in all cases.

$(6.2\text{-Å Co/7.7-Å Pt})_n$ multilayer films with repeat number $n = 5$ and 10 were prepared by sputtering on Si substrate with 22-Å Pt protection layer. The multilayer structure of Co/Pt multilayers was confirmed by a low angle X-ray diffraction and the extended X-ray absorption fine structure analysis. Both films exhibit a perpendicular magnetic anisotropy.

2. Results and Discussion

Static hysteresis curves (black) were determined by static MOKE measurement, as seen in Fig. 1. The coercivity ($H_c$) in case of $n = 5$ is larger than in case of $n = 10$, implying a larger perpendicular magnetic anisotropy, which is also confirmed by the anisotropy constant measurement. For $n = 5$ and 10, the magnetic anisotropy constant are $6.34 \times 10^6$ and $2.76 \times 10^6$ erg/cm³, respectively. Hysteresis curves (red) measured by stroboscopic pump-probe technique are also plotted together. The stroboscopic measurement was done at delay time of -2 ps. It is
clearly observed that, even at the delay time before the time-zero, the hysteresis curves measured by static and stroboscopic way become quite different, indicating the existence of the irreversible process effect from metastable magnetic states in the stroboscopic measurement. In case of n = 5, the reduction of $H_c$ from the stroboscopic measurement is vividly observed, while $H_c$ is quite similar to each other in case of n = 10. The slight change of loop shape for the case of n = 10, particularly in the nucleation region, might imply the existence of the irreversible process around this region.

Hysteresis curves measured by TR-MOKE at various delay times (t) are plotted for the cases of n = 5 and 10 in Fig. 2, where an external magnetic field ($H$) was swept from -1.7 to 1.7 kOe at each delay time. Examples of the hysteresis curve snapshot at t = 300 fs are also plotted. Compared to the case of Fig. 1 (t = -2 ps), $H_c$ even decreases further for both cases. For instance, in case of n = 10, $H_c$ decreased from 410 (t = -2 ps) to 278 Oe (t = 300 fs).

We have analyzed TR-MOKE signal following the increasing branch (red) of the hysteresis curve of Fig. 2(b) and (d). In Fig. 3(a), the TR-MOKE signal for n = 5 is plotted for various external fields from -1.0 to +1.0 kOe. It is observed that the TR-MOKE signal is quite symmetric under the inversion of the external field direction. TR-MOKE signal normalized by the peak value is plotted in Fig. 3(b), where most of curves collapse either upper (bluish) or bottom one (reddish). The black line in Fig. 3(a, b, d, and e) are Kerr signal at zero field. The intermediate TR-MOKE signal in green represents a case of external fields near $H_c$. It is considered that the TR-MOKE behavior is not significantly changing with respect to the external field, having a similar photoinduced demagnetization/remagnetization behavior for all fields. In Fig. 3(c), coercivity values determined from stroboscopic hysteresis curve are plotted with respect to the delay time. It should be noted that the coercivity from the stroboscopic pump-probe technique with pump-beam modulation ($H_{c, \text{pump}}$) denotes the pump-induced magnetization change. As seen in the figure, $H_{c, \text{pump}}$ seems to be almost the same within the error in case of n = 5.

TR-MOKE signal for n = 10 is plotted in Fig. 3(d). Overall, the trend looks similar to the case of n = 5. However, there is a clear difference in the details, for instance, under negative fields, where the TR-MOKE signal with remagnetization after the maximal demagnetization crosses zero-line from bottom (reddish) to upper region (bluish) for several intermediate external values, which is better seen if the signal is normalized as in Fig. 3(e). As seen in the figure, in case of 0.24 kOe external field, the TR-MOKE signal first increases sharply then
decreases, and again increases crossing from the bottom to the upper region. Considering that the TR-MOKE signal by pump-beam modulation is proportional to the photo-induced change ($\Delta M$) of magnetization, the observed behavior at the external field of 0.24 kOe implies that the magnetization, initially at negative region (reddish), experiences ultrafast demagnetization, followed by fast remagnetization reaching negative dip within 0.5 ps. Then this unusual remagnetization decreases, crossing the zero-line and moving upward, implying demagnetization on a time scale much longer than 5 ps. The corresponding $H_{c,\text{pump}}$ behavior with time is plotted in Fig. 3(f), where the similar nontrivial behavior is clearly observed. The unusual behavior is observed only when the positive (negative) external field is comparable to the coercivity while the net negative (positive) magnetization is excited by the pump beam pulse. Thus, it is considered that the unusual behavior arises from the complex magnetic configuration potentially with multiple magnetic domain structures. For further investigation, magnetic domain structures and their time-resolved behavior should be studied, which is beyond the scope of the present paper. We urge an experimental exploration of the ultrafast domain observation during the photo-induced demagnetization/remagnetization process for a specific external field condition around the coercivity of the sample.

We have carried out TR-MOKE measurement for a longer time scale up to 700 ps time delay, as in Fig. 4. In a similar way to the case of Fig. 3, TR-MOKE signal under several magnetic fields, normalized TR-MOKE signal, and $H_{c,\text{pump}}$ are plotted on a longer time scale. Overall, the field-dependent TR-MOKE behavior as well as the normalized one exhibits similar behavior on the longer time scale. It should be reminded that the Co/Pt multilayer in the present study has the PMA and thus, the TR-MOKE signal is free from the spin precession since the field is applied normal to the film plane and the probe is also detecting polar-MOKE signal. As seen in Fig. 4, spin precession does not significantly affect the observed TR-MOKE behavior, as expected. In case of $n = 5$, $H_{c,\text{pump}}$ (Fig. 4(c)) exhibits almost the same trend, compared to the case of the shorter time scale (Fig. 3(c)). In case of $n = 10$, the $H_{c,\text{pump}}$ keeps increasing after 100 ps, which might be ascribed to the characteristics of the irreversible behavior.

We have further quantitatively analyzed hysteresis parameters as in Fig. 5(a), where determination of the saturation magnetization ($\Delta M_s$), remanence ($\Delta M_r$), coercivity ($H_{c,\text{pump}}$), and hysteresis area ($\Delta A_{\text{pump}}$) from the TR-hysteresis are exemplified. In Fig. 5(b), $H_{c,\text{pump}}$ vs. delay time is plotted for $n = 5$ and 10 cases for a longer time scale up to 700 ps (open dot). For $n = 5$, $H_{c,\text{pump}}$ exhibits a rather simple behavior, mostly remaining below 0.1 kOe, as expected.
from the case of Fig. 3(c). However, it is interesting to see that $H_{C,pump}$ in case of $n = 10$ never remains constant over 700 ps. In the earlier phase, $H_{C,pump}$ is observed to be greater than 0.25 kOe, then abruptly decreases reaching the minimum at $t \sim 50$ ps. For $t > 50$ ps, $H_{C,pump}$ monotonically increases up to 700 ps. Considering that the $H_{C,pump}$ behavior is closely involved with the unusual $\Delta M$ behavior, as seen in Fig. 3(e) and (f), it is expected that the unusual $\Delta M$ behavior lasts over several hundreds of ps. It should be reminded that the coercivity behavior could be rather complex, particularly in ultrafast photo-induced demagnetization/remagnetization dynamics since there might be a multiple domain configuration involved with ultrafast dynamics. However, our experimental finding implies that, even around the coercivity region with possibly complex domain configuration, $H_{C,pump}$ could be still a stable parameter, characterizing the unusual dynamical behavior. In case of $n = 5$, where irreversible process is quite obvious, $H_{C,pump}$ remains almost the same. In case of $n = 10$, $H_{C,pump}$ exhibits a quite unusual behavior consistent with the nontrivial $\Delta M$ behavior determined from TR-MOKE signal, which suggests that the $H_{C,pump}$ could be also the stable description parameter for the photo-induced demagnetization/remagnetization dynamics regardless of reversible/irreversible categorization. Lastly, we have compared $H_{C,pump}$ and $\Delta A_{pump}/\Delta M_S$, where it is interesting to note that almost the exact matching exists for the two in both $n = 5$ and 10 cases. This is expected from the conventional static hysteresis loop analysis. For instance, for the square loop, the coercivity is determined by (hysteresis area)/(saturation magnetization). It is clearly observed that the matching in the static case also exists even in the case of ultrafast TR-hysteresis behavior.

3. Conclusions

We have investigated magnetic hysteresis curves measured by pump-probe stroboscopy technique adopting TR-MOKE on an ultrafast time scale. We have observed unusual hysteresis behavior together with the nontrivial $H_{C,pump}$ variation. For Co/Pt multilayer film with perpendicular magnetic anisotropy, irreversible and reversible trend in hysteresis curves are found for different multilayer repeat numbers. In both irreversible and reversible cases, $H_{C,pump}$ is closely related to the overall spin dynamics during the photo-induced ultrafast demagnetization/remagnetization process.
Acknowledgments

This research has been supported in part by Global Research Laboratory Program [Grant No 2009-00439] and by Max Planck POSTECH/KOREA Research Initiative Program [Grant No 2016K1A4A4A01922028] through the National Research Foundation of Korea (NRF) funded by Ministry of Science, ICT & Future Planning. This study was supported by Korea Research Foundation (NRF) grant No. 2018R1A2B3009569 and a Korea Basic Science Institute (KBSI) Grant D39614. This work was supported by Korea Institute for Advancement of Technology(KIAT) grant funded by the Korea Government(MOTIE) (P0008763, The Competency Development Program for Industry Specialist).
References


Figures

**Figure 1** Static hysteresis curve (black) and stroboscopically measured hysteresis curve (red) at delay time $t = -2$ ps for $n = 5$ and 10 case.

**Figure 2** Stroboscopically measured hysteresis curve (a) 3D-map for different delay times in case of $[\text{Co/Pt}]_5$ and (b) Snapshot at $t = 300$ fs. (c) 3D-map for different delay times in case of $[\text{Co/Pt}]_{10}$ and (d) snapshot at $t = 300$ fs.

**Figure 3** In case of $[\text{Co/Pt}]_5$ multilayer, (a) TR-MOKE signal for various fields from -1.0 to +1.0 kOe, (b) normalized TR-MOKE signal, and (c) $H_{c_pump}$, coercivity determined from the pump-beam modulation in stroboscopically measured hysteresis curve at 5 ps. In case of $[\text{Co/Pt}]_{10}$ multilayer, (d) TR-MOKE signal for various field from -1.0 to +1.0 kOe, (e) normalized TR-MOKE signal. The case of 0.24 kOe field value is denoted. (f) $H_{c_pump}$, coercivity determined from the pump-beam modulation in stroboscopically measured hysteresis curve at 5 ps. The horizontal dotted line represents the case of 0.24 kOe. The vertical dotted line s indicate the region of interest.

**Figure 4** $[\text{Co/Pt}]_5$ multilayer: (a) TR-MOKE signal for various fields from -1.0 to +1.0 kOe, (b) normalized TR-MOKE signal, and (c) $H_{c_pump}$, coercivity determined from the pump-beam modulation in stroboscopically measured hysteresis curve at 700 ps. $[\text{Co/Pt}]_{10}$ multilayer: (d) TR-MOKE signal for various field from -1.0 to +1.0 kOe, (e) normalized TR-MOKE signal. The case of 0.24 kOe field value is denoted. (f) $H_{c_pump}$, coercivity determined from the pump-beam modulation in stroboscopically measured hysteresis curve on a longer time scale (~ 700 ps)

**Figure 5** (a) Example of the determining pump-induced coercivity ($H_{c_pump}$), remanence ($\Delta M_R$), hysteresis area ($\Delta A_{pump}$), and saturation magnetization ($\Delta M_S$). The hysteresis area is colored red. (b) Comparison between $H_{c_pump}$ (open dot) and $\Delta A_{pump}/\Delta M_S$ (solid line) with respect to time for $[\text{Co/Pt}]_5$ (black) and $[\text{Co/Pt}]_{10}$ (red) multilayer.
Figure 1.
Figure 2

[Co/Pt]_5

(a)

[Co/Pt]_{10}

(c)

(b) t = 300 fs

(d) t = 300 fs
Figure 3
Figure 4
Figure 5