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Suppression of all-optical switching in He^+ -irradiated Co/Pt multilayers: influence of the domain-wall energy

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Abstract. The observation of all-optical helicity-dependent switching (AO-HDS) in ferri- and ferro-magnetic materials depends not only on the saturation magnetization, but on the magnetic domain size which takes into account the competition between the demagnetizing energy and the domain wall (DW) energy. In this paper, we study

the optical response of Co/Pt multilayers for which perpendicular magnetic anisotropy can be tuned using He⁺-irradiation. This allows us to correlate the observation of AO-HDS with the magnetic properties of the He⁺-irradiated multilayer. These results give direct evidence of the crucial role of the DW energy in the laser-induced switching process.

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The manipulation of magnetization using ultrashort laser pulses and without any other stimuli has triggered huge interest as an emergent field of research [1, 2]. This all-optical switching (AOS) of magnetization was initially demonstrated in ferrimagnetic GdFeCo alloy films using single femtosecond (fs) laser pulses [3]. This laser-induced switching in GdFeCo alloys was shown to be purely thermal [4] and ultrafast, taking place on a picosecond timescale [5]. A helicity-dependent switching was also obtained in these ferrimagnetic alloys but only for a narrow range of fluence [6], and its microscopic origin is still heavily debated [6, 7]. Later, AOS was also demonstrated in a much larger variety of ferrimagnetic alloy films and multilayers [8, 9] and also in ferromagnetic materials and spintronic devices [10, 11, 12, 13]. In contrast to GdFeCo alloy films, AOS in these materials was shown to be helicity-dependent independently on the laser fluence [9], cumulative and only achieved after a helicity-independent multiple-domain formation [14].

Furthermore, recent studies have investigated the magnetic parameters ruling the observation of all-optical helicity-dependent switching (AO-HDS) in these material classes [15, 17, 16, 18]. Indeed, El Hadri *et al* [18] performed a comprehensive investigation on ferrimagnetic TbCo alloy films and ferromagnetic Co/Ni and Co/Pt multilayers for a wide range of composition and thickness. By taking into account the competition between the demagnetizing energy and the domain wall (DW) energy, they showed that the large magnetic domain size is a criterion for the observation of AO-HDS, which is common for both studied ferri- and ferro-magnets [18]. More importantly, such investigation reconciles the contradictions between the reported low remanence criterion for ferrimagnetic materials [16, 15, 17] and the observation of AO-HDS in ferromagnets with a high saturation magnetization [10]. However, this investigation has mainly elucidated material classes where both the demagnetizing energy and the DW energy change [18], which makes it impossible to reveal the direct role of the DW energy in the AO-HDS process.

To elucidate the direct role of the DW energy in the observation of AO-HDS, we present in this paper an experimental investigation of the AO-HDS ability in a new material class, namely He^+ -irradiated Co/Pt multilayers with perpendicular magnetic anisotropy (PMA). Indeed, it has already been shown by Chappert *et al* in their pioneering work [19] that the PMA in ferromagnetic Co/Pt multilayers can be decreased in a controlled way via He^+ -ion irradiation. The irradiation process induces intermixing at the Co/Pt interfaces, which results in a strong decrease of the interface anisotropy and thus of the PMA and the DW energy [19, 21, 20, 22]. On the other hand, the saturation magnetization of the He^+ -irradiated Co/Pt multilayers remains constant [23]. Therefore, the change of the demagnetizing energy is negligible compared to the one of the DW energy in He^+ -irradiated Co/Pt multilayers. Hence, investigating the AO-HDS ability in He^+ -irradiated Co/Pt multilayers will enable to reveal the direct role of the DW energy in the AO-HDS process. In this context, we investigate the optical response

of He⁺-irradiated Co/Pt multilayers under the action of fs laser pulses. We demonstrate that a behavior change from AO-HDS to thermal demagnetization takes place by gradually increasing the irradiation dose. These findings highlight the direct role of the DW energy in the AO-HDS process, and confirm the requirement of a large domain size for the observation of AO-HDS.

In this paper, we decided to investigate the effect of He⁺-irradiation on the all-optical switching ability in Co/Pt multilayers which show whether pure AO-HDS or pure TD. On the other hand, it was previously demonstrated that as-deposited [Co (0.6 nm)/Pt (0.7 nm)]₂ and [Co (0.6 nm)/Pt (0.7 nm)]₄ multilayers show pure AO-HDS and pure TD, respectively [18]. Therefore, the investigated ferromagnetic samples are multilayer thin films of Glass/Ta(3 nm)/Pt(3 nm)/ [Pt(0.7 nm)/Co(0.6 nm)]_N/Pt(3 nm) grown by DC magnetron sputtering, where the number of repeats $N = 2$ or 4. The top Pt layer prevents sample oxidation. The studied films were subsequently irradiated with He⁺ ions at irradiation dose (ID) ranging from $5 \cdot 10^{14}$ He⁺/cm² to $5 \cdot 10^{15}$ He⁺/cm². The irradiation procedure was carried out at room temperature (RT) and at a constant energy of 15 keV. The changes in magnetic properties of He⁺-irradiated Co/Pt multilayers were measured by SQUID magnetometry. For none-irradiated [Co/Pt]_N multilayers, a saturation magnetization $M_s = 1619$ emu/cm³ (resp. 1982 emu/cm³) and a uniaxial anisotropy constant $K_u = 2.41 \cdot 10^7$ erg/cm³ (resp. $3.15 \cdot 10^7$ erg/cm³) were measured for $N = 2$ (resp. $N = 4$). Thus, the effective perpendicular anisotropy $K_{\text{eff}} = K_u - 2\pi M_s^2$ is positive for none-irradiated [Co/Pt]_N multilayers, which leads to a magnetization perpendicular to the film plane at zero fields. The increase of the saturation magnetization (M_s) with the number of repeats N is attributed to the spin polarization of Pt atoms by the adjacent Co layers [24]. The thickness of the spin-polarized Pt is not taken into account for the calculation of the M_s , and thus of the anisotropy constant (K_u).

For irradiated [Co/Pt]₂ multilayers, Fig. 1(a) shows that irradiation up to ID s of $5 \cdot 10^{15}$ He⁺/cm² results in square hysteresis at RT with the field oriented out-of-plane compared to the sample, indicating that all irradiated samples remain perpendicularly magnetized. The coercive field decreases from $H_c = 170$ Oe for the none-irradiated film to $H_c = 110$ Oe for films irradiated at $ID = 5 \cdot 10^{15}$ He⁺/cm². Moreover, Fig. 1(b) shows that irradiation up to ID s of $5 \cdot 10^{15}$ He⁺/cm² results also in a gradual decrease of the anisotropy field H_K . For irradiated [Co/Pt]₄ multilayers, Fig. 2(a) shows that irradiation up to 10^{15} He⁺/cm² also results in square hysteresis at RT with the field oriented out-of-plane compared to the sample. However, the magnetization easy axis is rotated towards the in-plane orientation for [Co/Pt]₄ multilayers irradiated at $ID > 10^{15}$ He⁺/cm², as depicted in Fig.2(b).

Fig. 3(a) shows that the anisotropy constant K_u decreases strongly down to $K_u = 2.41 \cdot 10^7$ erg/cm³ at a high $ID = 5 \cdot 10^{15}$ He⁺/cm², while the saturation magnetization remains almost constant. These findings are in agreement with previous studies on Co/Pt

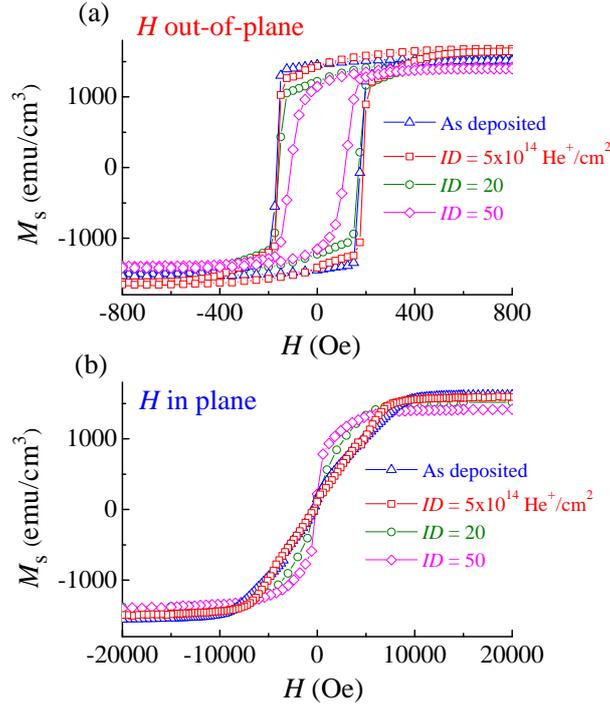


Figure 1. Room temperature SQUID hysteresis loops of the studied $[\text{Co}(0.6 \text{ nm})/\text{Pt}(0.7 \text{ nm})]_2$ multilayers as deposited, and after irradiation with 15 keV He^+ -ions at irradiation doses up to $5 \cdot 10^{15} \text{ He}^+/\text{cm}^2$. (a) (resp. (b)) The applied magnetic field is oriented out-of-plane (resp. in plane) compared to the sample. Lines are guides to the eyes.

multilayers [23, 25]. A similar trend is measured for irradiated $[\text{Co}/\text{Pt}]_4$ multilayers. As depicted in Fig. 3(b), K_u strongly decreases down to $2.17 \cdot 10^7 \text{ erg}/\text{cm}^3$ at $ID = 10^{15} \text{ He}^+/\text{cm}^2$, while M_s slightly decreases down to $1710 \text{ emu}/\text{cm}^3$.

On the basis of our previous study on the domain size criterion for AO-HDS [18], we estimate the magnetic domain size D for the He^+ -irradiated Co/Pt multilayers using the model proposed by Kooy and Unz [26] of periodic stripe domains with a strong uniaxial anisotropy in an infinite plate. We make the approximation that the domain wall size is negligible compared to the magnetic domain size and that the magnetization is perpendicular to the film plane within the domains. Therefore, the magnetic domain size D corresponding to the minimization of the total energy is expressed by:

$$D = t \exp \left[\frac{\pi D_0}{2t} + \ln \pi - 1 + \mu \left(\frac{1}{2} - \ln 2 \right) \right] \quad (1)$$

Where $D_0 = \frac{\sigma}{E_d}$ is the dipolar length and $\mu = 1 + \frac{E_d}{K_u}$ is the magnetic susceptibility. $E_d = 2\pi M_s^2$ is the magnetizing energy per unit volume, t the magnetic thickness, while A_{ex} is the exchange constant and $\sigma = 4\sqrt{A_{\text{ex}}K_u}$ is the DW energy per unit surface area. According to Eq. (1), the strong reduction of K_u for He^+ -irradiated Co/Pt multilayers

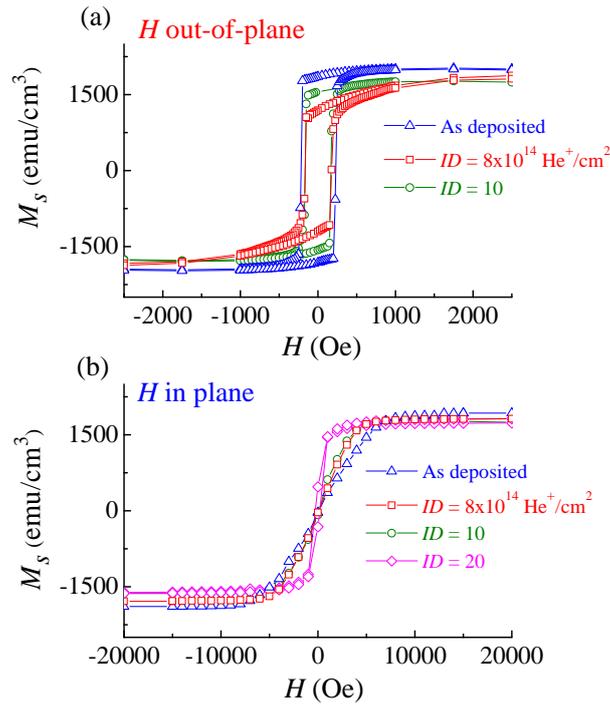


Figure 2. Room temperature SQUID hysteresis loops of the studied $[\text{Co}(0.6 \text{ nm})/\text{Pt}(0.7 \text{ nm})]_4$ multilayers as deposited, and after irradiation with 15 keV He^+ -ions at irradiation doses up to $2 \cdot 10^{15} \text{ He}^+/\text{cm}^2$. (a) (resp. (b)) The applied magnetic field is oriented out-of-plane (resp. in plane) compared to the sample. Lines are guides to the eyes.

at high ID s leads to a decrease of the DW energy, and thus of the domain size D . In a first approximation, we suppose that M_s and A_{ex} change only weakly with the irradiation dose. Therefore, we use the values of A_{ex} and M_s for none-irradiated samples in order to calculate the domain size for the He^+ -irradiated ones. The value of A_{ex} for none-irradiated Co/Pt samples is estimated using data from literature, $A_{ex}(\text{Co/Pt}) = A_{ex}(ID = 0) = 1.2 \cdot 10^{-6} \text{ erg/cm}$ [27]. We define a domain size threshold $D_{th} = 50 \mu\text{m}$ corresponding to the size of the fs laser spot used in the all-optical switching experiment (FWHM = $50 \mu\text{m}$).

Fig. 4(a) and Fig. 4(b) show the evolution of D as a function of the total magnetic thickness t for $[\text{Co}(\frac{t}{2})/\text{Pt}(0.7 \text{ nm})]_2$ and $[\text{Co}(\frac{t}{4})/\text{Pt}(0.7 \text{ nm})]_4$ multilayers, respectively. For $[\text{Co/Pt}]_2$ irradiated at $ID = 5 \cdot 10^{15} \text{ He}^+/\text{cm}^2$, we measured an anisotropy constant K_u ($ID = 5 \cdot 10^{15} \text{ He}^+/\text{cm}^2$) = $1.25 \cdot 10^7 \text{ erg/cm}^3$. As depicted in Fig. 4(a), the estimated domain size D for $[\text{Co}(0.6 \text{ nm})/\text{Pt}(0.7 \text{ nm})]_2$ multilayers with K_u ($ID = 5 \cdot 10^{15} \text{ He}^+/\text{cm}^2$) = $1.25 \cdot 10^7 \text{ erg/cm}^3$ decreases and coincides with the laser spot size D_{th} .

Note that it was shown in previous experimental investigations that He^+ -irradiated of Co/Pt multilayers results in a strong reduction of the Curie temperature (T_c) [20, 21],

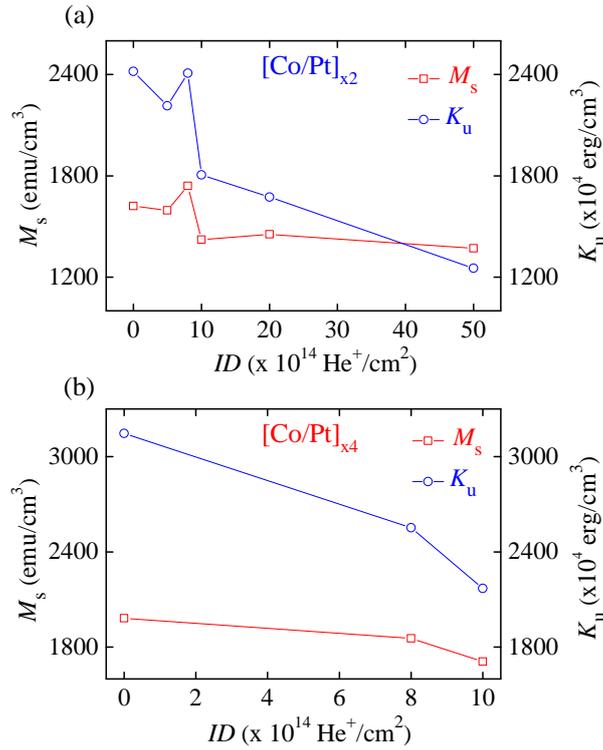


Figure 3. M_s and K_u dependence on the irradiation dose ID for the studied $[\text{Co}(0.6 \text{ nm})/\text{Pt}(0.7 \text{ nm})]_N$ multilayers with (a) $N = 2$ and (b) $N = 4$. Lines are guides to the eyes.

which is a hint that a decrease of A_{ex} might also take place. By taking into account a decrease of A_{ex} due to He^+ -irradiation, it can be understood from Eq.(1) that the domain size for $[\text{Co}(0.6 \text{ nm})/\text{Pt}(0.7 \text{ nm})]_2$ irradiated at $ID = 5 \times 10^{15} \text{ He}^+/\text{cm}^2$ will be smaller than the laser spot size D_{th} . On the other hand, recent experimental investigation showed that annealing can increase the exchange stiffness of ferromagnetic CoFeB films by 10% [28]. Hence, the He^+ -irradiation might lead to an increase of A_{ex} . To investigate the effect of an eventual increase A_{ex} , we estimated the domain size for $[\text{Co}(0.6 \text{ nm})/\text{Pt}(0.7 \text{ nm})]_2$ multilayers with K_u ($ID = 5 \times 10^{15} \text{ He}^+/\text{cm}^2$) = $1.25 \times 10^7 \text{ erg}/\text{cm}^3$ and with 10% and 20% increase of A_{ex} . Fig. 4(a) shows that the change in magnetic domain size with increasing A_{ex} by 10% is negligible.

According to the previously demonstrated domain size criterion for the observation of AO-HDS [18], $[\text{Co}(0.6 \text{ nm})/\text{Pt}(0.7 \text{ nm})]_2$ irradiated at $ID = 5 \times 10^{15} \text{ He}^+/\text{cm}^2$ are expected to exhibit thermal demagnetization (TD) and thus are different from non-irradiated $[\text{Co}(0.6 \text{ nm})/\text{Pt}(0.7 \text{ nm})]_2$ samples showing AO-HDS [10, 18]. Moreover, Fig. 4(b) shows that the domain size D for $[\text{Co}(0.6 \text{ nm})/\text{Pt}(0.7 \text{ nm})]_4$ calculated with K_u ($ID = 10^{15} \text{ He}^+/\text{cm}^2$) = $3.14 \times 10^7 \text{ erg}/\text{cm}^3$ is smaller than the laser spot laser. Therefore, irradiated $[\text{Co}(0.6 \text{ nm})/\text{Pt}(0.7 \text{ nm})]_4$ multilayers are also expected to exhibit TD.

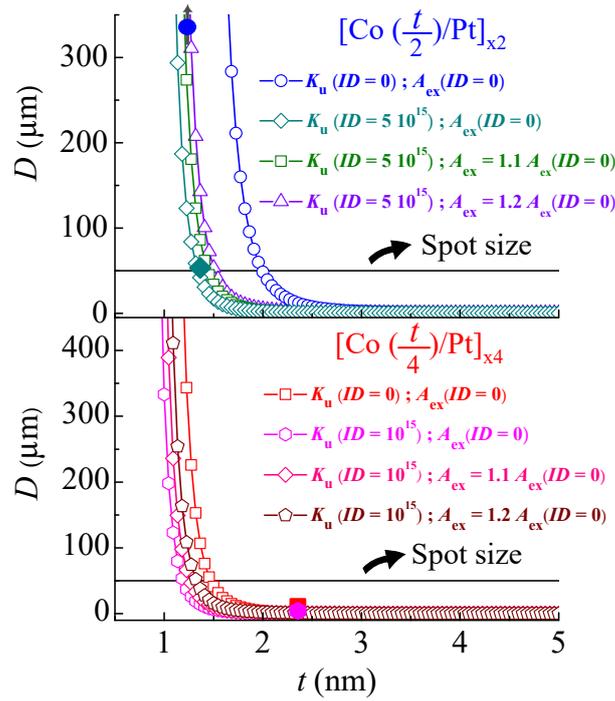


Figure 4. (a) Estimation of the magnetic domain size D as a function of the magnetic thickness t in $[\text{Co}(\frac{t}{2})/\text{Pt}(0.7 \text{ nm})]_2$ multilayers: for $K_u (ID = 0) = 2.41 \cdot 10^7 \text{ erg/cm}^3$ and $A_{ex}(ID = 0) = 1.2 \cdot 10^{-6} \text{ erg/cm}$; and for $K_u (ID = 5 \cdot 10^{15} \text{ He}^+/\text{cm}^2) = 1.25 \cdot 10^7 \text{ erg/cm}^3$ and three different values of A_{ex} [$A_{ex}(ID = 0)$, $1.1 A_{ex}(ID = 0)$ and $1.2 A_{ex}(ID = 0)$]. (b) Estimation of the magnetic domain size D as a function of the magnetic thickness t in $[\text{Co}(\frac{t}{4})/\text{Pt}(0.7 \text{ nm})]_4$ multilayers: for $K_u (ID = 0) = 3.14 \cdot 10^7 \text{ erg/cm}^3$ and $A_{ex}(ID = 0) = 1.2 \cdot 10^{-6} \text{ erg/cm}$; and for $K_u (ID = 10^{15} \text{ He}^+/\text{cm}^2) = 2.17 \cdot 10^7 \text{ erg/cm}^3$ and three different values of A_{ex} [$A_{ex}(ID = 0)$, $1.1 A_{ex}(ID = 0)$ and $1.2 A_{ex}(ID = 0)$]. D is calculated with M_s measured for the none-irradiated samples. The filled symbols indicate the magnetic thickness of the studied $[\text{Co}(0.6 \text{ nm})/\text{Pt}(0.7 \text{ nm})]_N$ multilayers.

In order to verify the expected behavior of He^+ -irradiated Co/Pt multilayers due to the action of fs laser beam, we used a Ti:sapphire fs laser with a 5-kHz laser pulse frequency, a wavelength of 800 nm, and a pulse duration of 35 fs. The Gaussian beam spot is focused with a FWHM of approximately 50 μm . The studied $[\text{Co}/\text{Pt}]_N$ samples are excited through the glass substrate with laser powers ranging from 0.75 mW for films with $N = 2$ up to 2.5 mW for films with $N = 4$. The response of the investigated magnetic films is probed using static Faraday microscopy in order to image the magnetic domains in transmission. Prior to being optically excited, the irradiated films are saturated with an external magnetic field. During the optical excitation, no magnetic field is applied and the 35-fs laser beam is swept from right to left for both circular polarizations with a low sweeping speed of approximately 10 $\mu\text{m/s}$.

We have first investigated the optical response of irradiated $[\text{Co}/\text{Pt}]_2$ multilayers

at ID ranging from $5 \cdot 10^{14} \text{ He}^+/\text{cm}^2$ to $5 \cdot 10^{15} \text{ He}^+/\text{cm}^2$. As depicted in Fig. 5, the none-irradiated ($ID = 0$) [Co/Pt]₂ multilayers exhibit AO-HDS, in agreement with the previous studies [10, 18]. By increasing ID from $5 \cdot 10^{14} \text{ He}^+/\text{cm}^2$ to $2 \cdot 10^{15} \text{ He}^+/\text{cm}^2$, the effect of the helicity on the irradiated [Co/Pt]₂ multilayers disappears gradually. Indeed, multiple domains start appearing in the scanned region and are stretched towards to the center of the beam spot. This behavior is attributed to thermal-induced DW motion [29, 30]. More importantly, Fig. 5 shows that only TD is obtained are for the [Co/Pt]₂ sample irradiated at $ID = 5 \cdot 10^{15} \text{ He}^+/\text{cm}^2$ and with a domain size D smaller than D_{th} (see Fig. 4(a)). Moreover, the multiple domains in the scanned region get smaller for $ID = 5 \cdot 10^{15} \text{ He}^+/\text{cm}^2$, thus confirming the reduction of the domain size due to the He⁺-irradiation. These findings confirm the large domain size criterion for the observation of AO-HDS [18], and give direct evidence that the DW energy plays a crucial role in the AO-HDS process. Furthermore, the lateral size of the laser-affected area for the studied [Co/Pt]₂ samples is expected to increase with the irradiation dose, since the He⁺-irradiation of [Co/Pt]₂ leads to a decrease of the Curie temperature [20, 21]. Hence, it can be seen from Fig. 5 that this lateral size increases by 28% for the [Co/Pt]₂ irradiated at $ID = 5 \cdot 10^{15} \text{ He}^+/\text{cm}^2$ compared to the none-irradiated [Co/Pt]₂ sample.

We have also verified the TD behavior of irradiated [Co/Pt]₄ multilayers under the optical excitation. As depicted in Fig. 6 the none-irradiated [Co/Pt]₄ multilayers show TD, which in agreement with the previous studies [10]. Moreover, Fig. 6 shows that the irradiated [Co/Pt]₄ multilayers with an estimated domain size $D < D_{th}$ exhibit also TD, as expected from the large domain size criterion [18]. Furthermore, the multiple domains in the scanned area of irradiated [Co/Pt]₄ multilayers become too small to be optically resolved, which also confirms the reduction of the domain size due to the He⁺-irradiation.

Recently, many experimental investigations and theoretical models have highlighted the microscopic origins of the symmetry breaking mechanism leading to AO-HDS in ferromagnetic materials. These origins can be divided into pure thermal origins such as the magnetic circular dichroism (MCD) [31, 32], and non-thermal origins such as the inverse Faraday effect (IFE)[33]. Our experimental investigation on He⁺-irradiated Co/Pt multilayers does not explain the symmetry breaking mechanism, but gives insights into the material-related conditions for the observation of AO-HDS. Indeed, the DW energy plays a crucial role in the AO-HDS process by impacting the magnetic domain size. The smaller is the DW energy, the smaller is the domain size. During the AO-HDS process, the initial effect of the laser pulses' helicity is suppressed in magnetic films with small DW energies via the formation of small magnetic domains during the cooling process.

On the other hand, it was previously reported in the literature that He⁺-irradiation influences the domain wall motion properties in ferromagnets, which could also explain the transition from AO-HDS to pure TD for the investigated He⁺-irradiated Co/Pt

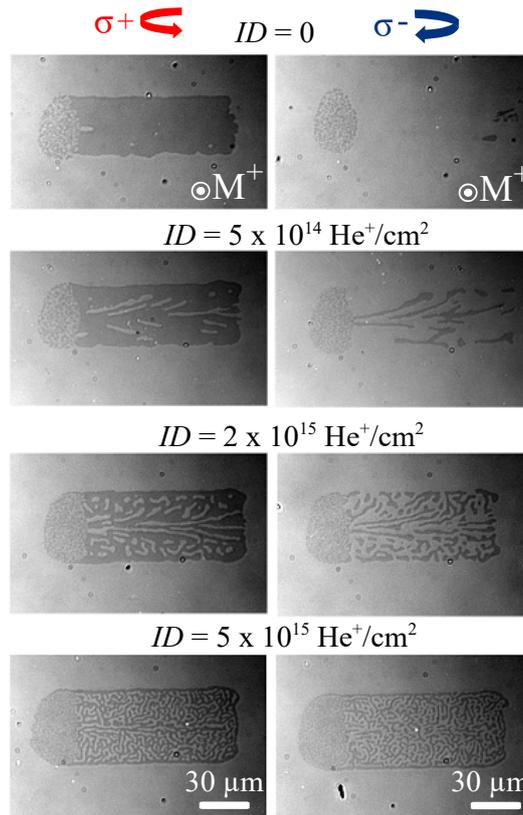


Figure 5. Optical response for $[\text{Co}(0.6 \text{ nm})/\text{Pt}(0.7 \text{ nm})]_2$ multilayers as a function of the irradiation dose ID under the action of swept 35-fs circularly polarized laser beam. A gradual transition from AO-HDS to pure TD is obtained by increasing ID up to $5 \cdot 10^{15} \text{ He}^+/\text{cm}^2$. For each sample, right- and left-circularly polarized laser beam were swept over the sample from right to left. The initial magnetization saturation up is exemplarily shown. The dark contrast corresponds to a reversal to down. The measurements were performed using a constant laser power of 0.75 mW.

multilayers. Indeed, Hererra Diez *et al* showed that the He^+ -irradiation of ferromagnetic CoFeB/MgO films with PMA leads to an increase of their depinning field in the creep regime [34]. Moreover, it was recently shown that the AO-HDS in ferromagnetic Co/Pt films is governed by a helicity-dependent domain wall motion-like mechanism [14, 32, 35]. Therefore, the He^+ -irradiation of ferromagnetic Co/Pt multilayers might also influence the helicity-dependent domain wall motion mechanism via the increase of the depinning field, thus canceling the helicity effect and leading to a transition from AO-HDS to TD regime.

In conclusion, we have experimentally investigated the AO-HDS ability in ferromagnetic He^+ -irradiated Co/Pt multilayers. We showed that increasing the irradiation dose leads to a strong decrease of the magnetic anisotropy, and thus of the magnetic domain size. More importantly, we demonstrated that a gradual change of behavior from AO-HDS to TD is observed in irradiated Co/Pt multilayers by gradually increas-

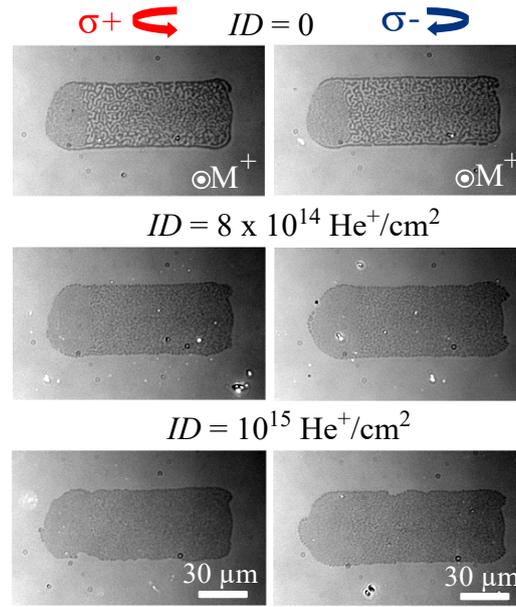


Figure 6. Optical response for $[Co(0.6 \text{ nm})/Pt(0.7 \text{ nm})]_4$ multilayers as a function of the irradiation dose ID under the action of swept 35-fs circularly polarized laser beam. A pure TD is obtained and the domain size in the laser excited decreases by increasing ID up to $10^{15} \text{ He}^+/\text{cm}^2$. For each sample, right- and left-circularly polarized laser beam were swept over the sample from right to left. The initial magnetization saturation up is exemplarily shown. The dark contrast corresponds to a reversal to down. The measurements were performed using a constant laser power of 2.5 mW.

ing the irradiation dose. Only TD is obtained for Co/Pt multilayers irradiated at high irradiation doses and with an estimated magnetic domain size smaller than the laser spot size. These findings confirm the large domain size criterion for the observation of AO-HDS and highlight the direct role of the DW energy in the AO-HDS process. Indeed, the initial effect of the laser pulses' helicity during AO-HDS process is suppressed in magnetic films with small DW energies via the formation of small magnetic domains during the cooling process. From a technological point of view, these findings elucidate the effect of the manipulation of PMA on the observation of AO-HDS in materials that are superior for data storage applications.

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