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All-optical helicity-dependent magnetic switching by first-order azimuthally polarized vortex beams

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In this letter, a method to realize all-optical helicity-dependent magnetic switching (AO-HDS) using a first-order azimuthally polarized vortex (FAPV) beam is demonstrated. Numerical calculations of the focal fields of FAPV beams reveal that left-handed and right-handed circular polarizations are generated due to the interaction between the polarization singularity and the helical wave front. Its feasibility for AO-HDS is experimentally demonstrated in $\text{Gd}_{27}\text{Fe}_{63.87}\text{Co}_{9.13}$ under low numerical aperture (NA) conditions and within a narrow fluence window. It is numerically predicted that under high NA conditions, the lateral size of magnetic bits recorded by FAPV beams can be nearly 30% smaller than that obtained by circularly polarized beams, which opens a promising route to realize ultrafast and ultrahigh-density magnetic recording. *Published by AIP Publishing.*

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The research upsurge into ultrafast magnetization dynamics was triggered by the observation of sub-picosecond demagnetization dynamics in nickel thin films after 60 fs pulsed-laser excitation¹ and the subsequent discovery of all-optical helicity-dependent magnetic switching (AO-HDS) in ferrimagnetic GdFeCo by circularly polarized (CP) light.² Initially, it was assumed that magnetization switching could only be accomplished with circular polarization. Later, purely thermal switching by linearly polarized light was demonstrated^{3,4} and a lot of research into several hypotheses, including helicity-dependent effective magnetic fields via the inverse Faraday effect (IFE),^{2,5,6} magnetic circular dichroism (MCD),⁷ and exchange coupling between the rare earth (RE) and transition metal (TM) sub-lattices³ was done in an attempt to explain the observed helicity-dependent and helicity-independent phenomena. Among these opto-magnetic phenomena, AO-HDS with deterministic direction information is of interest for applications in modern magnetic storage as well as spintronics.

Usually, AO-HDS in ferrimagnetic GdFeCo occurs within a narrow window of pulse energies of the incident light. No switching and helicity-independent switching is achieved below and above this window, respectively. AO-HDS has also been demonstrated in other ferrimagnetic materials (TbCo ,⁸ TbFe ,^{9–12} DyCo ,¹³ HoFeCo ,¹³ etc.),

engineered RE/TM multilayers, RE-TM heterostructures,¹³ and even in ferromagnetic materials (Co/Pt multilayers, FePtAgC granular films, etc.).¹⁴ In summary, helical electromagnetic fields of incident femtosecond pulses play a significant role in light-matter interactions and the subsequent AO-HDS in a wide variety of opto-magnetic materials.^{15–17}

In addition to helical polarization distributions, helical wave fronts or phase distributions carried by vortex beams are of paramount importance to light-matter interactions as well.^{18–21} It has been shown that circular polarization with high purity within the focal region can be generated by focusing first-order azimuthally polarized vortex (FAPV) beams, due to the interaction between the incident polarization singularity and the helical wave front of light.²² Its combination with opto-magnetic control has led to purely longitudinal magnetization needles²² and spherical magnetization chains^{23,24} with diffraction-limited feature sizes via the IFE. Furthermore, the focal spot size of FAPV beams can be smaller than that of CP beams under high numerical aperture (NA) conditions,^{25,26} which holds great potential for high density AO-HDS. However, although a number of promising prospects of FAPV beams have been theoretically predicted, their practical implementations with opto-magnetic materials are yet to be demonstrated.

In this letter, we experimentally demonstrate that under low NA conditions, FAPV beams can reproducibly reverse the magnetization direction in ferrimagnetic $\text{Gd}_{27}\text{Fe}_{63.87}\text{Co}_{9.13}$ in a

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helicity-dependent manner. A window of pulse energies of about 1.8% for the AO-HDS is revealed. It is numerically shown that under high NA conditions, this AO-HDS process promises nearly 30% smaller recorded magnetic bits compared with those achieved by ordinary CP beams.

According to the Debye diffraction theory,^{27–30} the focal electric fields of FAPV beams can be expressed as

$$\mathbf{E}(r, \varphi, z) = \begin{bmatrix} \mathbf{E}_r \\ \mathbf{E}_\varphi \\ \mathbf{E}_z \end{bmatrix} = A l^m e^{im\varphi} \int_0^\alpha \begin{bmatrix} V_1 \\ iV_2 \\ 0 \end{bmatrix} e^{ikz \cos \theta} \sqrt{\cos \theta} \sin \theta d\theta, \quad (1)$$

where

$$V_1 = J_{m-1}(kr \sin \theta) + J_{m+1}(kr \sin \theta), \quad (2)$$

$$V_2 = J_{m-1}(kr \sin \theta) - J_{m+1}(kr \sin \theta). \quad (3)$$

Here, A is a constant and m , which equals ± 1 , is the topological charge of the helical phase. r , φ , z , and k are the cylindrical coordinates and the value of the wave vector in the focal space, respectively. J_{m-1} and J_{m+1} denote Bessel functions of the first kind. α is the converging angle corresponding to the radius of the incident beam. Equation (1) is represented by cylindrical vector components and indicates that the incident purely azimuthal polarization is transformed into radial and azimuthal polarization components in the focal region due to the mutual interaction between the incident polarization singularity and the helical phase of light. Moreover, a phase difference of $\pi/2$ exists between the radial and azimuthal components, which means that circular or elliptical polarizations are generated in the focal region.

Figure 1(a) shows the calculated normalized electric-field distributions in the focal plane. The wavelength is

400 nm and the NA equals 0.005. Left-handed and right-handed circular polarizations are generated by focusing FAPV beams with $m = 1$ and $m = -1$, respectively. In order to reduce the lateral size of the focal spot as much as possible, annular incident beams³¹ are commonly introduced besides increasing the NA of the focal objective. Figure 1(b) shows the dependence of the lateral size of the focal spots on the NA and the annular factor (β) of the incident FAPV beams, where β is defined as the ratio of the inner and outer radii of the incident annular beam. In Fig. 1(b), the upper and the lower surfaces represent the full width at half maximum (FWHM) of the focal spots of CP and FAPV beams, respectively. It can be seen that under high NA conditions ($\text{NA} > 0.7$),²⁷ the focal spot of the FAPV beam is smaller than that formed by the CP beam and moreover this tendency turns more pronounced with higher NA and larger values of β . Figure 1(c) shows the focal size reduction versus NA and β by focusing FAPV beams compared to CP beams. Specifically, when $\text{NA} = 0.95$ and $\beta = 0.95$, the lateral focal sizes of the CP and the FAPV beams are 228 nm and 160 nm, respectively, which corresponds to a focal size reduction of nearly 30%, making FAPV beams interesting for nanoscale magnetic switching.

Figure 1(d) shows the schematic setup to demonstrate this helicity-dependent prediction. We used GdFeCo, which was well studied in AO-HDS, in our experiments. The amorphous ferrimagnetic $\text{Gd}_{27}\text{Fe}_{63.87}\text{Co}_{9.13}$ sample was grown by magnetron sputtering in a multilayer structure: glass/AlTi (10 nm)/SiN (5 nm)/GdFeCo (20 nm)/SiN (60 nm), where the AlTi layer served as a heat sink and SiN was used as a buffer and capping layer. Thin films of this alloy usually exhibit strong perpendicular magnetic anisotropy, and have a Curie point $T_C \approx 550$ K, a magnetization compensation point $T_M \approx 474.5$ K, and an angular momentum compensation point

FIG. 1. (a) Calculated normalized electric-field distributions in the focal plane by focusing FAPV beams with a wavelength of 400 nm ($\text{NA} = 0.005$). Left-handed and right-handed circular polarizations are generated in the focal region when $m = 1$ and $m = -1$, respectively. (b) Dependence of the lateral focal size on the NA and the annular factor β . The upper and the lower surfaces represent the FWHMs of the focal spots of CP and FAPV incident beams, respectively. (c) The focal size reduction versus NA and β by focusing FAPV beams compared with the focal size achieved by CP beams. (d) Schematic illustration of the setup to realize AO-HDS by focusing a FAPV beam.

$T_A \approx 524.5$ K.^{32–34} The magnetic domain structures were imaged through the magneto-optical Faraday effect of a linearly polarized pulsed beam from an amplified Ti:sapphire laser. The repetition rate, pulse width, and the central wavelength of the imaging laser pulses were 1 kHz, 40 fs, and 800 nm, respectively. Magnetic domains with magnetization parallel (“up”) or antiparallel (“down”) to the sample normal are shown as white or black regions, respectively, in the image of a CCD camera. The sample was excited by a single laser pulse or sweeping a pulse train with a central wavelength of 400 nm, which was obtained through frequency doubling of the regeneratively amplified 800 nm pulses by a BBO crystal. A pulse picker was used to reduce the repetition rate of the excitation pulses. The polarization of the excitation pulses was converted to azimuthal polarization by an ARCoTix polarization converter. A helical phase plate with topological charges of ± 1 , namely a helical $0-2\pi$ phase plate, was utilized to generate the FAPV beam. The experiments were performed at room temperature in air.

To verify the feasibility of AO-HDS in $\text{Gd}_{27}\text{Fe}_{63.87}\text{Co}_{9.13}$ by FAPV beams without ambiguity, we performed experiments with a low NA focal lens ($\text{NA} = 0.005$) which focused the incident laser beam down to a spot with a FWHM of around $42 \mu\text{m}$. Figure 2(a) shows the initial magnetization structure before the excitation of the incident light. The scale bar is $50 \mu\text{m}$. The results achieved by the excitation of the FAPV beams with $m = 1$ and $m = -1$ are shown in Figs. 2(b) and 2(c), respectively. In Figs. 2(b) and 2(c), the dots were written by single laser pulses with a fluence of 5.08 mJ/cm^2 on the sample. The fluence is defined as the pulse energy divided

FIG. 2. (a) The initial magnetization structure before light excitation. The white and black regions represent magnetization distributions with orientations “up” (M^+) and “down” (M^-), respectively. (b) The results achieved by the excitation of a FAPV beam with $m = 1$. The dots are generated by single laser pulses and the line structure is formed through sweeping an incident pulse train with a repetition rate of 100 Hz. (c) The results achieved by the excitation of a FAPV beam with $m = -1$.

FIG. 3. Helicity-dependent and helicity-independent switching formed by laser pulses with different fluences. (a) Combinations between the helicity of the incident FAPV beam and the initial state of the magnetization. The scale bar is $10 \mu\text{m}$. (b) Helicity-independent switching generated by two consecutive laser pulses with a fluence of 5.51 mJ/cm^2 . Annular switching patterns are finally formed in the regions where the conditions of AO-HDS are satisfied. (c) Helicity-dependent switching formed by a single laser pulse with a fluence of 5.08 mJ/cm^2 .

by the beam area which is measured through the Liu method.³⁵ It can be seen that AO-HDS occurs under this light fluence. In the case of $m = 1$, small black domains are recorded in the white region while nothing occurs in the black region. The opposite happens when $m = -1$. This AO-HDS is further demonstrated by sweeping the FAPV pulsed beams with a repetition rate of 100 Hz across the domain wall between the white and black regions. The sweeping speed of the beam was around $29.8 \mu\text{m/s}$. Similar to the case of single pulse excitation, helicity-dependent line structures are formed in the corresponding regions. In contrast, with a higher light fluence, helicity-independent switching occurs. Figure 3(a) shows the

FIG. 4. Switching probability as a function of the fluence of the incident FAPV single pulses with different topological charges. The blue hollow and the red solid squares with error bars represent the switching probabilities under the excitation of the FAPV single pulses with $m = 1$ and $m = -1$, respectively. The data are fitted with an error function. The magnetization initially orients “up”.

FIG. 5. Switching areas obtained under the excitation of FAPV beams ($m = 1$) with different fluences. The minimum switching diameter is around $1.1 \mu\text{m}$ at a fluence of 4.83 mJ/cm^2 . No switching is observed when the fluence is lower than 4.83 mJ/cm^2 .

combinations between the helicity of the incident FAPV beam and the initial state of the magnetization. As shown in Fig. 3(b), under the excitation of two consecutive laser pulses with a fluence of 5.51 mJ/cm^2 , magnetic switching happens regardless of the helicity of the imposed phase or the initial state of the magnetization. Notably, as the light fluence at the rim of the switching region is lower than that in the center, annular switching patterns are finally formed in the regions where the conditions of AO-HDS are satisfied. Figure 3(c) shows the AO-HDS generated by a single laser pulse with a fluence of 5.08 mJ/cm^2 . These phenomena occur from the fact that all-optical magnetic switching in GdFeCo alloys dominantly stems from the exchange interaction between the Gd and Fe sublattices via a transient ferromagnetic-like state owing to the substantially different opto-magnetic dynamics of the Gd and Fe sublattices.^{3,4} Furthermore, AO-HDS occurs only in an appropriate fluence range of the exciting pulses, which is ascribed to the MCD.⁷ Above this range, helicity-independent switching would occur since both the left-handed and right-handed circularly polarized light can exceed the threshold switching fluence.

The switching probability as a function of the fluence is shown in Fig. 4. The initial magnetization was in the “up” direction. This procedure was repeated about 20 times at every pulse energy. The blue hollow and the red solid squares with error bars represent the switching probabilities under the excitation of the FAPV single pulses with $m = 1$ and $m = -1$, respectively. The experimental data are fitted with an error function.⁷ It can be seen that a distinct fluence window (Δ) of AO-HDS of around 1.8% is observed, which is calculated by

$$\Delta = \frac{F_{m=-1} - F_{m=1}}{\frac{1}{2}(F_{m=-1} + F_{m=1})}, \quad (4)$$

where $F_{m=1}$ and $F_{m=-1}$ are defined as the threshold switching fluences of $\text{Gd}_{27}\text{Fe}_{63.87}\text{Co}_{9.13}$ excited by the FAPV beams with $m = 1$ and $m = -1$, respectively. The switching probability is 0.5 at the corresponding threshold fluence. This narrow window induced by the MCD indicates that AO-HDS occurs in an appropriate fluence range of the exciting pulses. Beyond this window, no switching or helicity-independent switching occurs, which is similar to the previous results achieved by purely CP beams.⁷ The slope of the probability curves originates from pulse to pulse intensity fluctuations.

So far, AO-HDS excited by the FAPV beams has been experimentally demonstrated in $\text{Gd}_{27}\text{Fe}_{63.87}\text{Co}_{9.13}$ under both single-pulse and multi-pulse conditions. The switching

areas obtained under the excitation of FAPV beams ($m = 1$) with different fluences are shown in Fig. 5. It can be seen that the switched area turns smaller with lower excitation fluence of the FAPV beam. However, the smallest switching diameter obtained in our experiment was limited to around $1.1 \mu\text{m}$ at a fluence of 4.83 mJ/cm^2 , even by using focal lenses with increased NA. No switching was observed when the fluence was lower than 4.83 mJ/cm^2 . This relatively large switching diameter is due to the fact that the GdFeCo films are magnetically soft, limiting the smallest achievable domain sizes to about $1\text{--}3 \mu\text{m}$.³⁶ Although GdFeCo is therefore not optimal for demonstrating magnetic recording on the nanoscale, these results can inspire future developments to apply FAPV beams to other ferrimagnets (such as TbFe, TbCo, and TbFeCo)^{8–12,37} and ferromagnets (Co/Pt multilayers, FePtAgC granular films, etc.)¹⁴ which possess stable nanoscale magnetic domains.

In conclusion, in our work, FAPV beams are introduced to realize AO-HDS. By reversing the wave front helicity of the FAPV beams, deterministic AO-HDS is unambiguously observed in $\text{Gd}_{27}\text{Fe}_{63.87}\text{Co}_{9.13}$ under low NA conditions. A fluence window of about 1.8% for AO-HDS is obtained. Numerical calculations predict that compared with CP beams, a switching size reduction of nearly 30% can be achieved by using FAPV beams under high NA conditions, which enables magnetic switching of nanoscale domains. Even though the large domain size of the magnetically soft GdFeCo films restricts the demonstration of nanoscale magnetic switching in this material, ferromagnets and other ferrimagnets with nanoscale domain sizes may take full advantage of FAPV beams and be applied to realize nanoscale magnetic switching. The results in our work open a promising route to realize ultrahigh-density AO-HDS and may inspire subsequent developments in closely related fields, including data storage and spintronics.

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