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

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In this letter, we experimentally demonstrate that under low numerical aperture (NA) conditions, FAPV beams can reproducibly reverse the magnetization direction in ferrimagnetic Gd27Fe63.87Co9.13 in 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 Gd27Fe63.87Co9.13 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-demagnetization dynamics in nickel thin films after 60 fs dynamics was triggered by the observation of sub-picosecond ferrimagnetic GdFeCo by circularly polarized (CP) light.2 optical helicity-dependent magnetic switching (AO-HDS) in (MCD),7 and exchange coupling between the rare earth inverse Faraday effect (IFE),2,5,6 magnetic circular dichroism ing helicity-dependent effective magnetic fields via the 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 chains23,24 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 Gd27Fe63.87Co9.13 in a
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, the focal electric fields of FAPV beams can be expressed as

$$\mathbf{E}(r, \varphi, z) = \begin{bmatrix} E_r \\ E_{\varphi} \\ E_z \end{bmatrix} = A e^{im\varphi} \begin{bmatrix} V_1 \\ iV_2 \\ 0 \end{bmatrix} e^{ikz \cos \theta} \sqrt{\cos \theta} \sin \theta d\theta,$$

where

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

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

Here, $A$ is a constant and $m$, which equals $\pm 1$, is the topological charge of the helical phase. $r$, $\varphi$, $z$, 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. $\alpha$ 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 beams 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 ($\beta$) of the incident FAPV beams, where $\beta$ 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 ($NA > 0.7$), 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 $\beta$. Figure 1(c) shows the focal size reduction versus NA and $\beta$ by focusing FAPV beams compared to CP beams. Specifically, when $NA = 0.95$ 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 Gd$_2$Fe$_{63.87}$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 $T_M < T_C$. 

**FIG. 1.** (a) Calculated normalized electric-field distributions in the focal plane by focusing FAPV beams with a wavelength of 400 nm (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 $\beta$. 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 $\beta$ 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.
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 ARCoptix polarization converter. A helical phase plate with topological charges of ±1, namely a helical 0–2π 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 Gd27Fe63.87Co9.13 by FAPV beams without ambiguity, we performed experiments with a low NA focal lens (NA = 0.005) which focused the incident laser beam down to a spot with a FWHM of around 42 μm. Figure 2(a) shows the initial magnetization structure before the excitation of the incident light. The scale bar is 50 μm. 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/cm² on the sample. The fluence is defined as the pulse energy divided by the beam area which is measured through the Liu method. 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 μm/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. 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. 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 orients “up” initially.
Diameter (µm)

<table>
<thead>
<tr>
<th>Fluence (mJ/cm²)</th>
<th>10.6</th>
<th>7.2</th>
<th>4.2</th>
<th>2.8</th>
<th>1.1</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.40</td>
<td>5.25</td>
<td>5.10</td>
<td>4.95</td>
<td>4.83</td>
<td>&lt; 4.83</td>
</tr>
</tbody>
</table>

The probability curves originate from pulse to pulse intensity fluctuations. The probability is 0.5 at the corresponding threshold 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. It can be seen that a distinct fluence window (Δ) of AO-HDS of around 1.8% is observed, which is calculated by

\[
\Delta = \frac{F_{m=1} - F_{m=-1}}{\frac{1}{2}(F_{m=1} + F_{m=-1})},
\]

where \(F_{m=1}\) and \(F_{m=-1}\) are defined as the threshold switching fluences of Gd\(_{27}\)Fe\(_{63.87}\)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. 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 Gd\(_{27}\)Fe\(_{63.87}\)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 µm at a fluence of 4.83 mJ/cm², even by using focal lenses with increased NA. No switching was observed when the fluence was lower than 4.83 mJ/cm². 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 1–3 µm. 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) and ferromagnets (Co/Pt multilayers, FePtAgC granular films, etc.) 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 Gd\(_{27}\)Fe\(_{63.87}\)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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