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Magnetic helicoidal dichroism with XUV light carrying orbital angular momentum

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Abstract. Circularly polarized light is a fundamental tool in magnetic studies, notably for magnetization dynamics. It is less common in magneto-optics to exploit the orbital angular momentum (OAM) of value ℓ carried by light beams possessing a helical wavefront. After finding many applications in the visible range, recently OAM pulses with ultra-short duration and XUV wavelengths became available, widening the range of experiments that can be envisaged. We modelled the interaction of an XUV OAM beam with non-uniform magnetic structures, showing that the far field scattered intensity profile encodes the symmetry of the magnetic structure in a way that depends on the sign and value of ℓ . In analogy with magnetic circular dichroism, this effect, named magnetic helicoidal dichroism (MHD), can be observed by inverting the sign of either the orbital momentum or of the magnetization. We obtained experimental evidence of MHD by measuring ℓ -dependent resonant scattering from a magnetic vortex. The results of recent complementary experiments match well the theoretical predictions, confirming the potential of the new toolset provided by MHD for studying the laser-triggered ultrafast dynamics of complex magnetic materials.

1. Introduction

The interaction of polarized light beams, ranging from infrared to hard-x-rays, with magnetic materials defines the rich set of analytical tools used in magneto-optics and it is also widely employed for studying magnetization dynamics in time-resolved experiments. Photons carrying a spin angular momentum (SAM) $\sigma\hbar$, with $\sigma = \pm 1$ corresponding to left or right circular polarization (figure 1a), have been used



intensely for decades in circular dichroism experiments to discriminate and study the properties of magnetic, as well as of chiral, molecules and materials. It is less common, in magnetic dichroism studies, to exploit the fact that a photon beam can also carry an orbital angular momentum (OAM) with topological charge equal to $\ell\hbar$ (with $\ell \in \mathbb{Z}$), which corresponds to featuring a helical wavefront [1]. Such OAM beams, also called *light vortices*, develop a distinctive corkscrew shape of the wavefront (see figure 1b) determined by an azimuthal angular dependence of the electric field phase and show a well-defined handedness. The analogy between quantum mechanics and paraxial optics suggests that modes with such an azimuthal dependence are eigenvalues of the angular momentum operator L_z and carry an OAM of $\ell\hbar$ per photon [2].

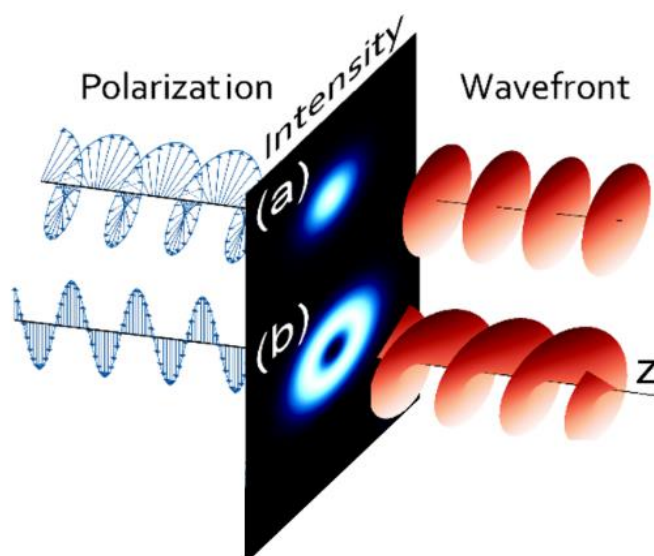


Figure 1. Sketches of SAM and OAM photon beams. On the left, blue arrows represent the electric field vectors defining the polarization, circular in (a) and linear vertical in (b). On the right, phase wavefronts are shown in orange for SAM (a) and OAM (b) photon beams. Blue maps on black background represent the intensity in a given transverse plane.

OAM beams in the visible range have been generated and intensely exploited for decades as a tool for manipulating physical systems and probing their properties [3,4]. More recently, OAM beams with shorter wavelengths, from the extreme ultraviolet (XUV) [5-7] to the soft X-ray [8] range, became available at high-harmonic generation (HHG) [5,6], free-electron laser (FEL) [7] and synchrotron [8] sources, widening considerably the range of experiments that can be envisaged. Moreover, HHG and FEL sources produce light pulses with femtosecond and attosecond durations, ideally suited for dynamic studies.

In this framework, it has been theoretically predicted that OAM beams should prompt novel kinds of dichroism experiments, paving the way for new spectroscopic tools in the fields of orbital physics and magnetism [9]. In particular, after the scattering of an OAM beam from magnetic structures featuring non-uniform magnetization (like magnetic vortices), it is expected that the far field intensity profile encodes the symmetry of the magnetization distribution in a way that depends on the sign and value of ℓ [10], giving rise to magnetic helicoidal dichroism (MHD). As for magnetic circular dichroism, MHD can be observed by inverting the sign of either the orbital momentum or of the magnetization, i.e. by switching the handedness of either the light vortex or of the magnetic vortex.

2. Experimental details

The first experimental demonstration of MHD at XUV wavelengths was reported recently by measuring the scattering of an OAM photon beam from a magnetic vortex [11]. In order to support these findings and to prepare for more complex time-resolved measurements in pump-probe mode, a new series of experiments was planned and performed.

A new set of permalloy dots was prepared by magnetron sputtering and lithography, using either ion-etching or lift-off methods. The shape of the dots was the same as in the original experiment, pacman-like and optimized by means of micromagnetic simulations [12] in order to form at remanence a stable magnetic vortex whose sense of rotation, either clockwise (CW) or counter-clockwise (CCW), could be deterministically defined by an external magnetic field pulse (see figure 2). With respect to the original 15 μm , their lateral size was varied from 5 to 20 μm , allowing for a trade-off between ease of alignment of the XUV beam (~ 3 μm FWHM) on the dot and ease of formation of a clean vortex (favoured for smaller sizes). Dots were prepared in four sizes and two orientations (π -rotated). They formed a 12×12 matrix of 144 dots, 100 μm spaced, on each Si substrate in order to have at one's disposal fresh new samples in situ, since in the previous experiment radiation damage was observed after ~ 12 hours of exposure to the FEL beam. Preliminary MOKE measurements showed that samples prepared by lift-off provided better performance in terms of vortex formation and were selected for the final measurements.

As in the original experiment [11], the light source was the FERMI FEL-1 free-electron laser [13], tuned to produce 52.8 eV photons, i.e. to match the Fe-3p core resonance, in order to enhance magneto-optical effects at XUV wavelengths. The scattering experiment was performed at the DiProI end-station [14], equipped with high-precision translation and rotation stages for the sample alignment, with an electromagnet capable of generating field pulses in excess of 500 Oe along any direction within the sample surface, and with a charge-coupled device (CCD) for two-dimensional (2D) imaging of the scattered intensity. Well-defined OAM values up to $\ell = \pm 3$ were imparted to the XUV beam by using specially designed spiral zone plates (SZPs, prepared at PSI - Villigen) that worked at the same time as focusing elements (all with the same focal distance) and as phase plates [7].

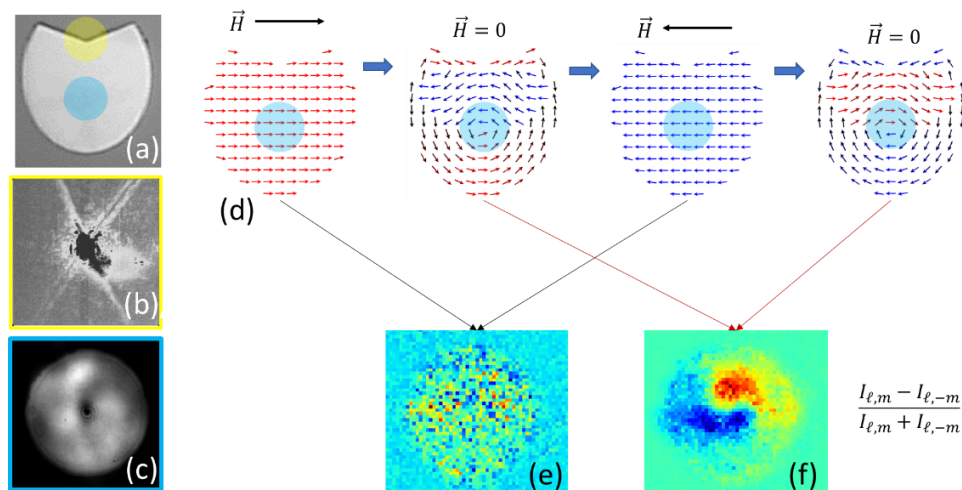


Figure 2. (a-c): Scanning electron microscopy image of a 15 μm wide permalloy dot (a). The colored circles correspond to the XUV beam size. Positioning the beam at the crossing of the straight edges produces a characteristic scattering pattern (b), used for aligning at the correct position for measurements (c). (d): Sequence of magnetic configurations prepared for each measurement (arrows represent the results of micromagnetic simulations). (e-f): magnetic signal obtained by combining two measurements performed in saturated (e) and remanent (f) conditions.

3. Results

A first part of the experiment was dedicated to designing a more reproducible procedure for aligning the photon beam at the desired position on the dots, corresponding to the nominal vortex center, since this point raised criticisms to our previous results. To do this, we took advantage of the intense scattering produced by the two straight edges of the pacman-shaped permalloy dot (see figure 2a,b), which make it possible to position precisely the beam at their intersection, then move the sample vertically to the desired position (figure 2c). The experiment was performed following a sequence of four measurements (figure 2d) for each sample and OAM value. The MHD signal was then obtained by taking the difference divided by the sum of two images of the scattered intensity collected either for opposite saturation directions (figure 2e) or for CW and CCW configurations of the magnetic vortex (figure 2f). These configurations were always obtained just by applying magnetic pulses, without any sample or SZP realignment in between the two acquisitions, minimizing possible spurious effects in the MHD signal.

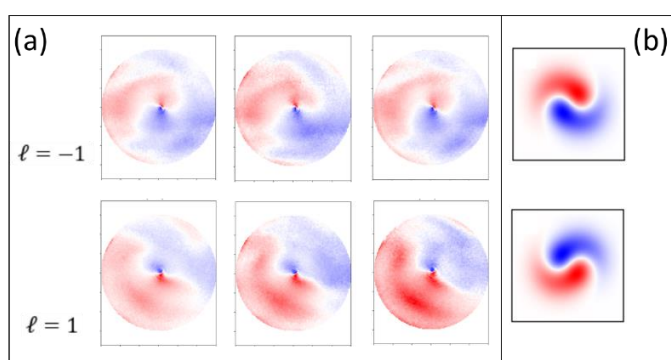


Figure 3. MHD signal measured (a) and calculated (b) in magnetic vortices for $\ell = -1$ (top) and $\ell = +1$ (bottom). The photon energy is 52.8 eV, the polarization is linear-p and the angle of incidence is 48 degrees. The three pairs of images in (a) correspond to three different permalloy samples.

Examples of data obtained using linear p-polarized radiation and a scattering angle close to the Brewster extinction condition are shown in figure 3a for $\ell = -1$ and for $\ell = +1$. The experimental results are compared to the calculated MHD intensity patterns (figure 3b) based on a previously developed theoretical model [10], showing an overall good agreement. In order to assess the reproducibility of the measurements and the reliability of the alignment procedure, we measured different dots, of either the same or of different size, prepared on different substrates, obtaining rather equivalent results (see figure 3a). Measurements on the smallest objects (5 μm) showed some scattering from the edges, hampering data analysis.

Finally, we tested also a setup for the pump-probe scheme. The 800 nm wavelength laser beam was made to arrive at the sample almost collinear with the FEL beam also in presence of the SZP and order-sorting aperture necessary for imparting the OAM to the incoming XUV radiation. Test measurements were performed, showing the expected demagnetization of permalloy after the laser pulse. The results are too preliminary for a full scientific discussion, but they demonstrate the feasibility of pump-probe experiments with XUV-OAM beams at DiProI.

4. Conclusion

The primary goal of our experiment was to confirm that magnetic structures can yield, in the XUV range, strong MHD signals, which depend on the exact topology of their magnetization configuration. We had obtained the first experimental evidence of MHD by measuring ℓ -dependent scattering from permalloy magnetic vortices at the FERMI FEL source [11]. We have now given support to our findings in a new series of more systematic experiments with results that match well our theoretical predictions [10] and confirm the potential of the new toolset provided by MHD for studying the laser-triggered ultrafast dynamics of complex magnetic materials. This novel effect has the potential to open up new perspectives for fundamental spectroscopic studies, both in the static and in the dynamic regimes, allowing for instance a dynamical mapping of the spin texture.

Understanding the interaction of OAM XUV-light with a complex magnetic structure has many potential interests. At a fundamental level, it allows to observe a new kind of dichroism, to study the role of photon spin-orbit coupling [15], as well as to explore new possibilities of angular momentum transfer between light and matter, as already observed in the visible range [3,16]. In terms of applications, MHD in reflection can be exploited as a new spectroscopic tool joining the family of magnetic dichroism techniques. For their symmetry and size, magnetic vortices can be considered as an ideal benchmark sample to explore the interaction with OAM beams; moreover, given their rich dynamical response in the ultrafast domain, they are promising structure for light-driven manipulation of the magnetization.

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