Magnetic Circular Dichroism probed using High Harmonics

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Abstract: We demonstrate the first generation and phase matching of circularly-polarized high harmonics, which are bright enough for magnetic circular dichroism measurements at the *M* absorption edges of the magnetic materials Fe, Co and Ni.
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Circularly polarized light in the extreme ultraviolet (EUV) and soft x-ray regions of the electromagnetic spectrum is extremely useful for exploring chirality-sensitive light-matter interactions. Magnetic circular dichroism (XMCD) makes it possible to extract detailed information about the magnetic state of matter and its interaction with phononic and electronic degrees of freedom on femtosecond time scales and nanometer length scales. Specifically, XMCD can be used to distinguish between spin and orbital contributions to the atomic magnetic moment in ferromagnetic materials, with element-specificity, which is not possible using ultrafast visible laser spectroscopy. However, to date, circularly polarized EUV and soft x-ray beams were restricted to large-scale electron storage facilities, such as synchrotrons and x-ray free electron lasers. Such facilities have great advantages of high peak and average powers in the x-ray region. However, drawbacks include limited access and temporal resolution, as well as ≈ 100 fs jitter.

Table-top short-wavelength sources based on high harmonic (HHG) up-conversion of femtosecond laser pulses represent a complementary and viable alternative to large scale sources, due to their unique ability to generate bright, broadband, ultrashort and coherent light from the UV to the keV region [1]. HHG sources have successfully exploited the resonantly enhanced magnetic contrast at the *M* absorption edges of the 3d ferromagnets Fe, Co and Ni in the EUV region to study element-specific dynamics in magnetic materials on femtosecond-to-attosecond time scales on the tabletop [2, 3]. This new capability has opened up a wealth of opportunities for greater fundamental understanding of correlated phenomena. Specifically, novel insights on ultrafast (few-femtosecond) nanoscale phenomena were uncovered by capturing spin scattering and transport, as well as exchange interaction dynamics, in complex multi-species magnetic materials [4-6]. However, these investigations have so far been limited to linearly polarized HHG: to date, generating circularly-polarized harmonics has been highly inefficient, reducing both the photon flux and degree of circularity to a level that precludes scientific applications [7-9].

In this work, we present the first direct approach for generating bright circularly-polarized HHG under phase-matching conditions [10], based on a technique that was suggested almost two decades ago [11,12] and recently measured [13]. Surprisingly, although for decades it was assumed that HHG from atoms was brightest when both the driving laser and HHG fields were linearly polarized, circularly-polarized HHG can be as bright as linearly-polarized HHG. We demonstrate the first magnetic circular dichroism measurements using a tabletop light source, in this case at the *M* absorption edges of Fe, Co and Ni. This experiment thus represents the first application of circularly-polarized HHG and the first measurement of the helicity of circularly-polarized harmonics.

In our experiment, we focus the fundamental and second harmonic of a Ti:sapphire laser amplifier with opposing helicities into a gas-filled hollow waveguide. As schematically depicted in Fig. 1, the polarization states of the two drivers can be independently adjusted in an interferometric Mach-Zehnder setup, thereby controlling the helicity of the generated harmonics. The threefold dynamical symmetry of the generation process gives rise to circularly polarized harmonics: the electric fields of harmonic orders $q = 3n+1$ have the same helicity as the fundamental, $q = 3n-1$ rotate with the second harmonic, while harmonic orders $q = 3n$ are completely suppressed. Using Ar, Ne and He gas, we produce a circularly-polarized HHG spectrum that spans the entire *M* absorption edges of the 3d ferromagnets, with a photon flux comparable to that used in previous ultrafast element-selective magneto-

optic HHG-based experiments [2-6]. Thus, the element specific magnetic state of materials can be probed simultaneously with spin and orbital selectively and with high signal-to-noise ratio, by exploiting XMCD.

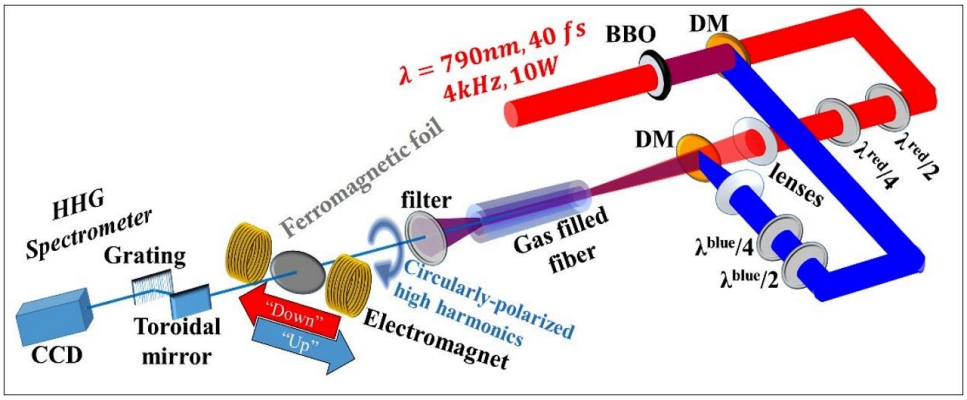

Figure 1 - Setup for generating circularly-polarized harmonics under phase-matching conditions.

Figure 2a plots HHG spectra from Ar at 90 Torr, in Ne at 650 Torr and in He at 900 Torr. Clearly, the $q = 3n$ harmonics are almost completely suppressed for all gas species. This feature is a clear indication that the HHG polarization is close to circular [13]. The detected photon flux is more than 10^9 photons per second. Next we measured the HHG spectrum after passing through a magnetized 50 nm thick Ni foil, and extracted the XMCD asymmetry by flipping the in-plane magnetization of the sample. Figure 2b presents the HHG spectrum when the foil is magnetized either "up" or "down", labeled by I^{up} and I^{down}, respectively. The normalized XMCD asymmetry, $A = (I^{up}-I^{down}) / (I^{up}+I^{down})$ is shown in Fig. 2c. The asymmetry of the $3n+1$ and $3n-1$ harmonics exhibit opposite signs, thereby proving that the helicity of the *3n+1* harmonics is indeed opposite to the *3n-1* harmonics. As the first bright source of circularly-polarized HHG, this work paves the way for investigating ultrafast circular dichroism of magnetic samples, chiral molecules and nanostructures.

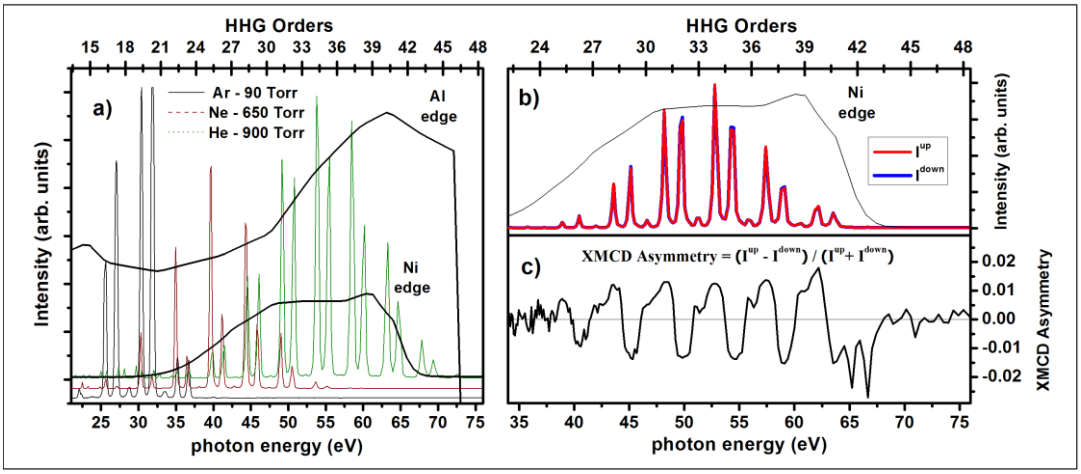

Figure 2 – a) Spectra of bright circularly-polarized HHG in Ar, Ne and He & b), c) XMCD of 50 nm Ni from HHG in He.

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