# Coherent x-rays driven by ultrashort-pulse lasers: generation, application, and prospects

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## ABSTRACT

Ultrashort laser pulses represent an ideal starting point for frequency conversion of light to almost any wavelength from the THz to x-rays. High-harmonic upconversion (HHG) is a unique process enabled by the combined strong field laser field and the few-cycle pulse duration of a femtosecond laser pulse. HHG makes it possible to generate coherent light in the spectral region from the vacuum-UV into the x-ray region at sub-nm wavelengths. HHG sources are now finding increasingly diverse application for both science and technology, in topics ranging from basic studies of atomic processes, to materials dynamics revealed through time and angle-resolved photoemission. Furthermore, the coherent nature of the HHG process makes possible unprecedented control over light in a new region of the spectrum, making it possible to, for example, control the polarization state and spectral bandwidth, creating the most complex time-domain waveforms ever measured and characterized. Here we review recent work, as well as efforts at commercial implementation of HHG sources.

**Keywords:** EUV, coherent diffractive imaging (CDI), high-harmonic generation

## 1. INTRODUCTION

By upconverting coherent ultrashort-pulse laser light to much shorter wavelengths through the high harmonic generation process, for the first time it is possible to implement what is essentially an “x-ray laser” light source on a tabletop. In HHG, coherent short-wavelength laser light is generated through the process of field ionization of an atom in a strong laser field. The full implications of this process are still being uncovered. Recent work, for example, has shown that (contrary to past expectations), the HHG process can generate very high-energy photons >>1 keV by driving the process with intense few-cycle mid-IR pulses,[1] as well as generating novel circularly-polarized harmonics.[2-4] The time and frequency structure of the HHG pulses is related; for example, when driven by mid-IR, the spectrum of HHG light merges into a supercontinuum that corresponds to a single attosecond pulse, while driving the process with deep-UV light generates narrow spectral peaks that correspond to a regular train of attosecond pulses with very little intrinsic chirp.[5]

Applications of coherent EUV HHG light have expanded greatly in recent years. This decade has seen continued advance in fundamental understanding and capabilities, allowing these new sources to be used for discovery science in a variety of areas, and most-recently attracting interest for possible use in industrial process metrology. Examples of experimental applications include observing the dynamics of the quantum exchange interaction fundamental to magnetic materials;[6-10] the use of coherent HHG light for tabletop nanoimaging with record resolution;[11-14] and studies of the physical limits of energy flow at the nanoscale.[15-17] Two examples are elaborated on below.

## 2. EXAMPLES

### 2.1 Coherent EUV Microscopy

The EUV region of the spectrum is well suited for imaging ICs because the short wavelengths allow for high resolution imaging. However, the EUV spectrum lacks suitable *refractive* lens elements, making it problematic to form images with high-resolution. To overcome this challenge, we use a lensless imaging technique known as ptychographic[18,19] coherent diffractive imaging (CDI).[20-22]

In ptychography, a coherent illumination beam is rastered across a sample, and the light scattered from the sample is recorded on a detector. The data is fed into an iterative phase retrieval algorithm, and a complex image of the sample is computationally reconstructed. The amplitude of the complex image yields information about the materials present on

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the sample, while the phase information primarily encodes topographical information. The resolution of the image is only limited by the wavelength of the illumination, lambda (λ), and the numerical aperture (NA) of the collected scatter patterns. In a recent work, images were obtained using 30nm illumination reflected from the sample, demonstrating 40nm (1.3λ) transverse and 0.6nm axial resolution[11] (see Fig.1). In a transmission geometry using 13.5 nm illumination, we demonstrated sub-15-nm resolution imaging.[23] In ptychography, the scanning is area-by-area rather than point-by-point, allowing data to be collected quickly, over large fields-of-view. Each scan position shares about 70% area overlap with adjacent scan positions (except for the areas at the edges of the scan). The overlap in scan positions provides redundant data allowing the ptychographic algorithm to converge quickly, compared with traditional single exposure CDI. Additionally, ptychographic algorithms can be extended to use partially coherent illumination, multiple colors, and orthogonal modes and polarization states.[13]

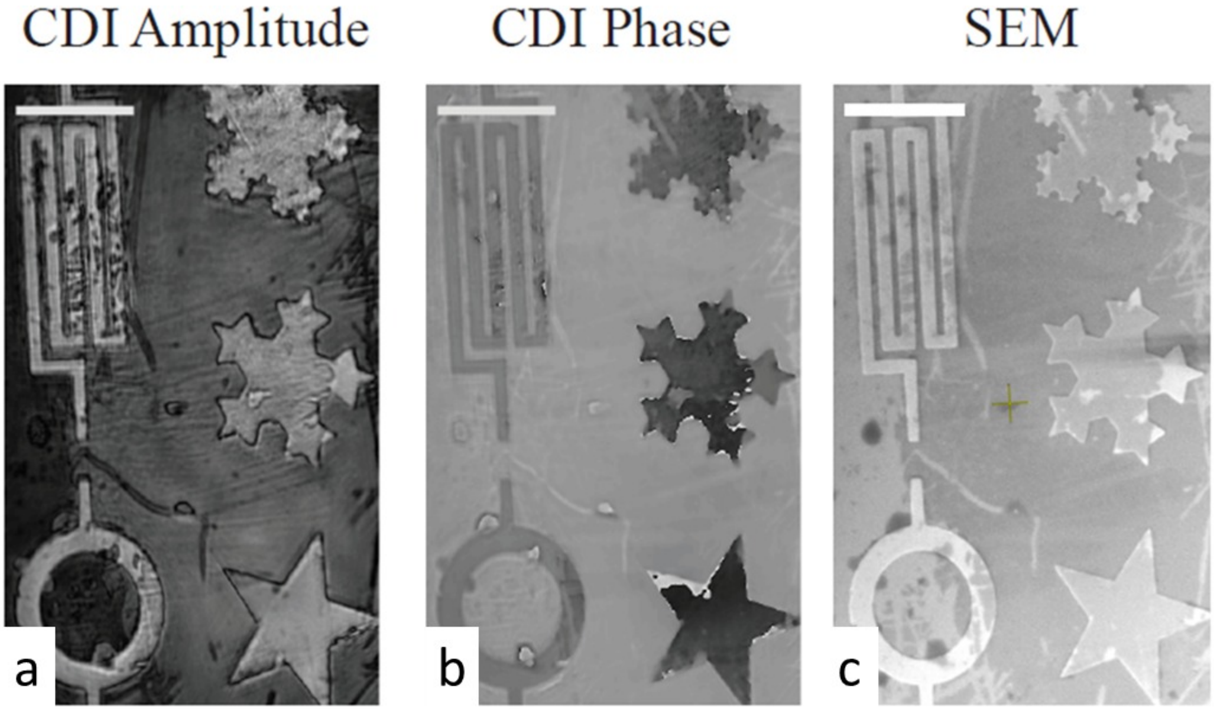

Figure 1. Nanoscale imaging of Ti features patterned on a Si wafer. The (a) amplitude and (b) phase of the ptychographic CDI images are shown to the left and center, respectively. For a comparison, a (c) SEM image of the same sample is shown on the right. With 30 nm illumination wavelength, the ~40 nm resolution represents diffraction limited resolution for the numerical aperture of the scattered light, in this case corresponding to ~42 nm. All the scale bars, upper left, correspond to 10µm. Figure adapted from Zhang *et al.*[11]

## 2.2 Element-specific dynamics in magnetic systems

The Magneto-optic Kerr Effect (MOKE) results in modulation of the reflectivity of a surface depending on the polarization of the incident light with respect to the direction of magnetization of the reflection surface. In the EUV, the MOKE asymmetry is greatly enhanced near the inner-shell absorption edges of the elements. Iron, Nickel, and Cobalt all have absorption edges in the photon energy range of 50-70 eV, making it possible to monitor magnetization in an element specific manner. Since HHG light is intrinsically short-pulse, this provides a method for monitoring dynamics in magnetic systems unique ways. Past work has demonstrated that, for example, in an alloy even through the elements are intermixed, the dynamics of demagnetization when a surface is heated by an ultrashort pulse, are decoupled with a time-energy scale corresponding to the exchange energy for the material.[9,10,24] In other work, element specificity was used as a method for tagging layers.[8] The data show that when a multilayer surface is heated with an ultrashort pulse, some of the observed demagnetization results from generation of a very large spin current into the bulk. This can be seen through the transient *increase* in magnetization of a buried Fe layer due to spin-polarized electron transport from the Ni overlayer.

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