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Fast spectroscopic imaging using extreme ultraviolet interferometry

Hannah C. Strauch[a], Fengling Zhang[b], Stefan Mathias[a], Thorsten Hohage[c], Stefan Witte[b,d], and G. S. Matthijs Jansen[a]

[a]1st Institute of Physics, University of Göttingen, Germany
[b]Advanced Research Center for Nanolithography ARCNL, Amsterdam, the Netherlands
[c]Institute for Numerical and Applied Mathematics, University of Göttingen, Germany
[d]Imaging Physics dept., Faculty of Applied Sciences, TU Delft, The Netherlands

ABSTRACT

Extreme ultraviolet pulses as generated by high harmonic generation (HHG) are a powerful tool for both time-resolved spectroscopy and coherent diffractive imaging. However, the combination of these techniques to achieve spatio-spectro-temporal data remains hardly explored due to the challenging and time-consuming data acquisition. Here, we present Fourier-transform spectroscopic holography (FTSH), an interferometric approach to spectroscopic imaging that combines Fourier-transform spectroscopy with holography. By encoding spectral information in the measured diffraction pattern, FTSH dramatically reduces the sampling requirements by one order of magnitude. This enables us to record full spectroscopic imaging data in less than 2 minutes, and makes FTSH especially promising for femtosecond time-resolved nano-spectroscopy using table-top HHG sources.

Keywords: Ultrafast dynamics, EUV, HHG, Imaging, Spectroscopy, Interferometry

1. INTRODUCTION

High-harmonic generation (HHG) is a unique light source for extreme ultraviolet (EUV) light, since it combines excellent spatial coherence with an ultrabroad spectrum and ultrashort pulse duration. This makes HHG a highly attractive platform for methods ranging from nanoscale coherent diffractive imaging to time-resolved spectroscopy. Importantly, HHG-based EUV spectroscopy enables both attosecond time-resolution and element-specificity, which enables to disentangle correlated electronic, structural and magnetic dynamics in condensed matter (e.g.,[1–3]). As a consequence, HHG-based methods that combine nanoscale spatial resolution with EUV spectroscopy are highly sought-after. However, the aim for spatio-spectro-temporal data places a significant requirement on the data acquisition.

Holography plays an important role in the development of advanced EUV microscopy. For example, time-resolved EUV microscopy is commonly performed using Fourier-transform holography (FTH) as a starting point,[4] and this approach has recently been extended with photon-energy scanning at a free-electron laser to image a structural phase transition with full spatio-spectro-temporal resolution.[5] For HHG light sources, though, the necessity of spectral filtering for such photon-energy scanning dramatically increases the experimental cost, making spectrally multiplexed imaging highly desirable. A potential solution is ptychography, which enables diffraction-limited resolution with multi-spectral illumination[6,7] through scanned illumination of the sample in overlapping steps. However, the need for overlap is significantly increased for multi-spectral illumination,[7,8] thereby challenging the acquisition of fully spatio-spectro-temporal resolved data.

Here, we present Fourier-transform spectroscopic holography (FTSH), an interferometric approach to spectroscopic imaging that combines Fourier-transform spectroscopy (FTS) with Fourier-transform holography.[9] FTSH enables us to record full spatio-spectral information using the full EUV bandwidth of our HHG light source in less than 2 minutes, and is therefore especially promising to realize femtosecond time-resolved spectroscopic imaging using table-top HHG sources.

Further author information:
G.S.M.J.: E-mail: gsmjansen@uni-goettingen.de

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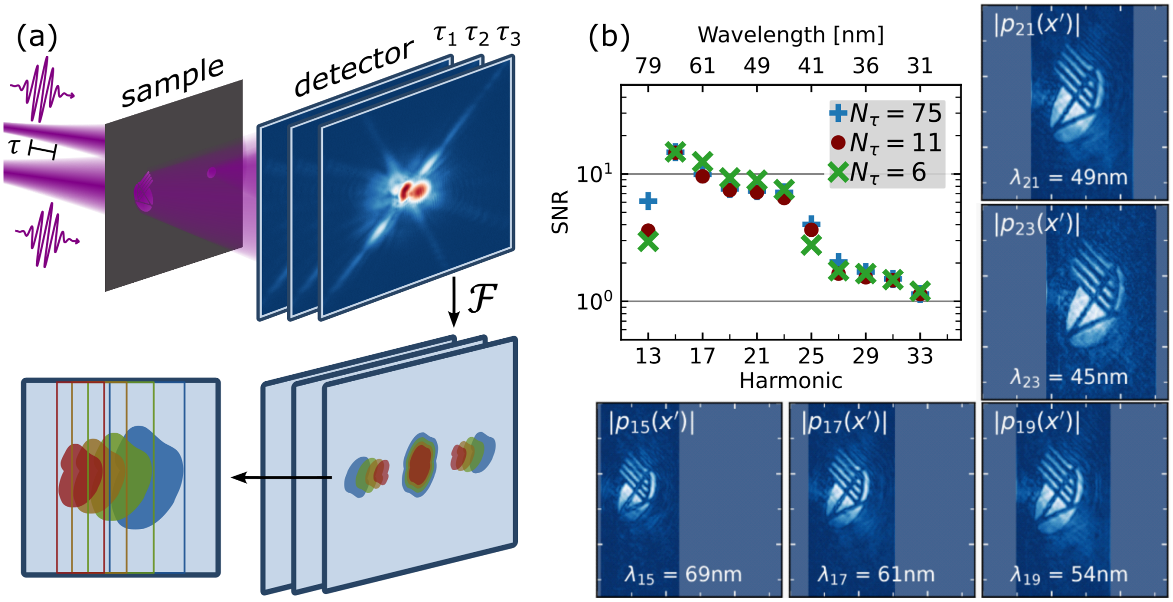

Figure 1. Schematic overview and exemplary results of Fourier-transform spectroscopic holography (FTSH). a) In a Fourier-transform holography setup, two coherent beams illuminate a sample and reference structure, giving rise to a far-field diffraction pattern from which an image of the sample can be retrieved with a single spatial Fourier transform. When multiple wavelengths are present in the illumination are present, this method yields partially overlapping holograms for each wavelength. Full spectroscopic imaging data can then ideally be achieved by joining holography with Fourier-transform spectroscopy, where the phase between the two illumination beams is controlled. b) Here, we show FTSH results achieved using a pair of extreme-ultraviolet high-harmonic generation light sources. The combination of holography and spectroscopy dramatically reduces the experimental sampling requirements, and compared to conventional Fourier-transform spectroscopy, FTSH enables fully multiplexed imaging with identical image quality at less than 10% of the experimental measurement duration.

2. METHODS AND RESULTS

The experimental method is outlined in Fig. 1a. In a FTH setup, we illuminate the object and reference structures using two independent phase-controlled EUV beams,[10] and record the resulting far-field diffraction patterns using an EUV-sensitive CCD camera. Like in conventional FTH, the resulting interference pattern can be analyzed in a single step using a spatial Fourier transform to calculate the hologram, which is the convolution of the transmitted waves after the object and reference. In this case, the broad EUV spectrum of our HHG source (spanning from 80 to 30 nm) leads to 11 partially overlapping holograms. To disentangle the full spectral information, we use FTS, where the relative delay between the illuminating EUV pulses is shifted.

In this approach, the measurement efficiency and duration is limited by the set of delays that is required to fully sample the temporal interference pattern. In conventional FTS (whether imaging or not), this is determined by the Nyquist-Shannon theorem. For HHG light, this implies that $2N_{\max}$ delay steps are necessary, where $N_{\max}$ is the order of the highest harmonic. In FTSH, however, a significant amount of prior knowledge is available that can be used to directly reduce the sampling requirement: 1. The wavelengths of the individual high harmonics are (usually) known a-priori. 2. The spatial Fourier transform that is used to analyze the FTH pattern yields complex-valued holograms that thus provide both amplitude and phase information simultaneously. 3. As sketched in Fig. 1a, the individual holograms are only partially overlapping, with no overlap at all between well-separated wavelengths.

From 1 and 2, it follows that the amplitude and phase of each spectral component can be determined from just N_λ carefully-chosen delay steps, where N_λ is the number of high harmonics present in the illumination (11 in our case). With 3 in addition, it follows that the number of measurements can be reduced even more (to 6 in this case), where the reduction in sampling requirement depends on both the bandwidth of the EUV spectrum and the geometry of the FTH sample. In Fig. 1b, exemplary imaging results are shown for the outlined minimal sampling strategy. Moreover, a comparison of the resulting image quality (in terms of signal-to-noise ratio) is presented for the three sampling strategies (conventional Nyquist-Shannon with $N_\tau = 75$ delay steps, using 1+2 with $N_\tau = 11$ delay steps, and using 1+2+3 with $N_\tau = 6$ delay steps). The measurement time per delay step was identical for all methods. Crucially, we find that each sampling strategy yields equivalent image quality - but at a dramatically reduced measurement duration for the proposed FTSH minimal sampling strategies.

In summary, FTSH explicitly encodes the EUV spectral information in the measured diffraction pattern and so yields a uniquely efficient spectroscopic imaging method. This is especially promising for time-resolved (pump-probe) spectroscopic imaging.

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