HHG-based EUV Beamlines for Spectroscopy and Coherent Diffractive Imaging

Nguyen Xuan Truong, Ilya Strashnov, and Melissa Denecke

School of Chemistry, The University of Manchester, Oxford Road, Manchester M13 9PL, United Kingdom

Giacomo Bertali and M. Grace Burke

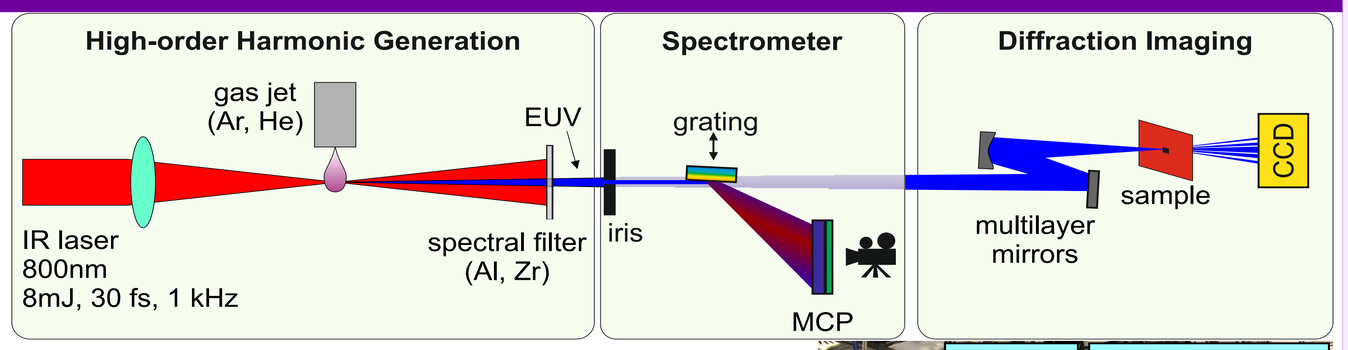
School of Materials, The University of Manchester, Oxford Road, Manchester M13 9PL, United Kingdom

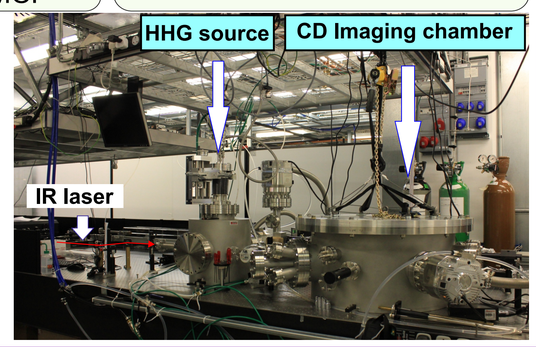
Introduction

High-order harmonic generation (HHG) provides novel coherent EUV radiations which are interesting for spectroscopy of atoms, molecules, bulk materials and coherent diffractive imaging (CDI) of nanosize materials. The coherent sources are nowadays achievable on a table-top scale, allowing to perform small lab experiments, which have normally been done in big facilities like synchrotrons or free electron lasers. We present a newly built HHG beamline in Manchester providing EUV light with the photon energy of up to 120 eV. We perform coherent diffractive imaging of (graphite) nanoparticles. A time-delay compensated monochromator (10-100 eV) will be set-up, which delivers sub 10 fs EUV pulses. This beamline is crucial for, e.g., time-resolved angular resolved photoemission spectroscopy (TRARPES) of metal oxides.

Experimental Methods

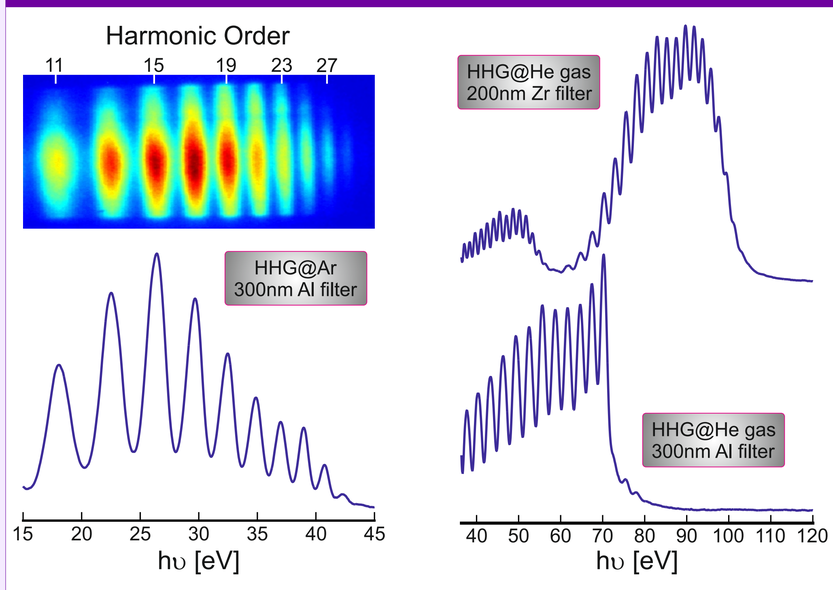
The driven Ti:sa laser system provides IR pulses with pulse energy of up to 8 mJ, duration of 30 fs at a central wavelength of 800nm and a 1 kHz repetition rate. To produce the harmonics, the IR beam is focussed with a lens into a 8 mm long gas-cell with a diameter of about 200 μm. Argon or helium gas at a backing pressure of 700 mbar is fed into the gas-cell with a piezo-driven pulsed valve (Attotech). The gas-cell is mounted on a precise R-XYZ manipulator for fine adjustments. The generated HHG beam is separated from the fundamental IR beam using spatial and spectral (Al/Zr) filters. To characterize the HHG, a home-built flat-field EUV spectrometer was used. The spectrometer consists of a 3mm x150μm iris, laminar-type replica diffraction gratings (300l/mm, 1200l/mm, and 2400l/mm), a microchannel plate (MCP) with a phosphor screen at the backside, and an air-side CCD camera. The absolute HHG yields are calibrated with a XUV photodiode or a soft X-ray CCD camera (PXO-2048B, Princeton Instruments).





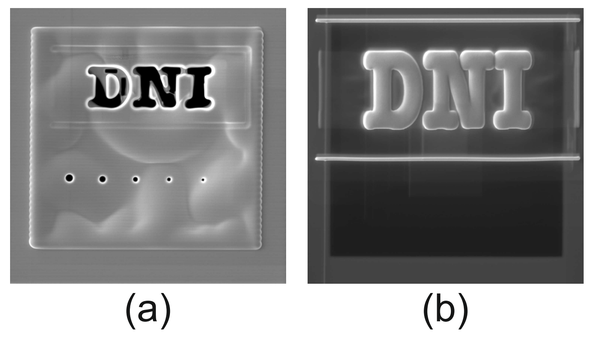
For CDI experiments, we use multilayer mirror pairs in a Z-configuration to select the HHG radiation at 30 nm and 13 nm and focus (f=100 and 500 mm) the beam on to a sample containing nanoparticles (see below for details). The diffracted image is oversampled with a vacuum soft x-ray CCD camera. With the help of phase retrieval algorithms [1], the contour structure of the particles can be determined.

Results



- HHG spectra generated with argon and helium gases. With argon, the HHG spectrum peaks around 15-60 eV, depending on the experimental conditions (laser pulse energy, gas pressure, etc.). With helium, a spectral cutoff around 100 eV is observed. An approach to extend the cutoff photon energy is to employ longer driven laser wavelengths (>1.5 μm). To enhance the HHG yield, two color (800nm + NIR) HHG methods can be applied.

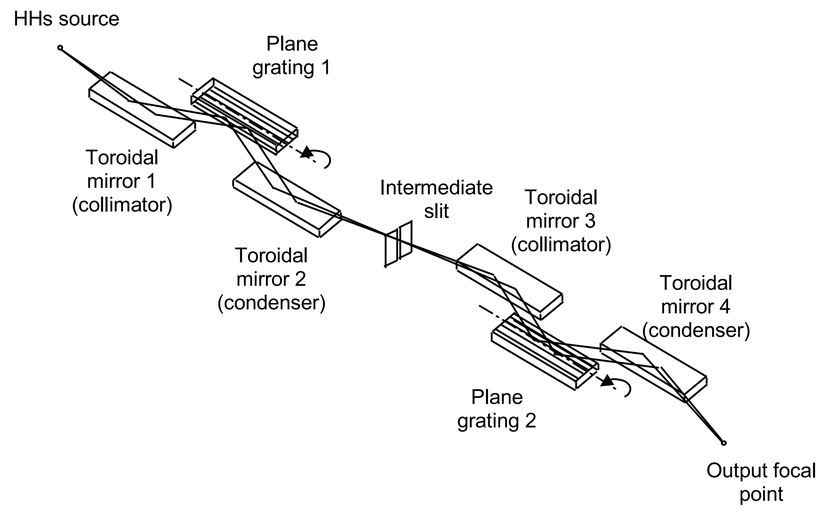
- CDI Test Samples Preparation and Characterization.
Samples are prepared on Si chips with a 10μmx10μm window (Norcada Inc, Canada), covered by a 20nm thick silicon nitride membrane. An FEI Helios NanoLab 660 DualBeam FIB was used to prepare site-specific structures within the SiN window at nanometric level through either selective milling (SE FEG-SEM image (a)) or Pt sputtering (BSE FEG-SEM (b)).



Outlook

- **Time-delay-compensated monochromator for HHG based EUV sources**

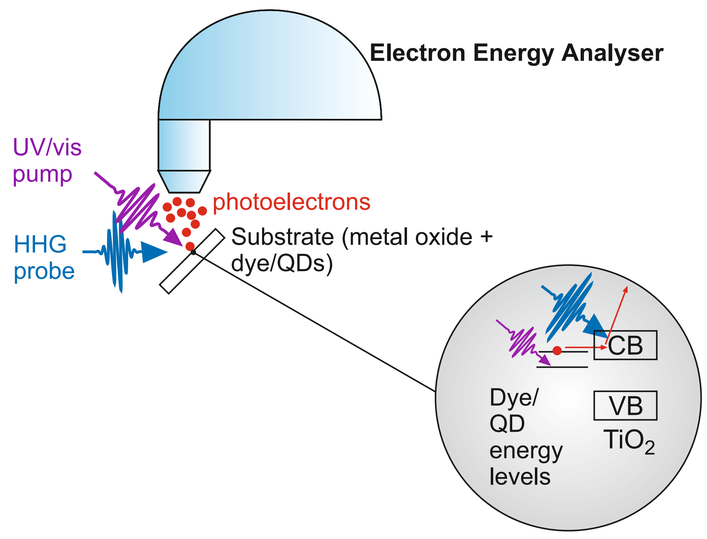
The monochromator contains four toroidals and a set of grating pairs, designed by Poletto and coworkers [2]. It covers the spectral range of up to 100 eV with an energy resolution of 300 meV and temporal resolution of 10 fs. (collaborations with L. Poletto and F. Frassetto, Italy)



- **Femtosecond laser - mass spectrometry**

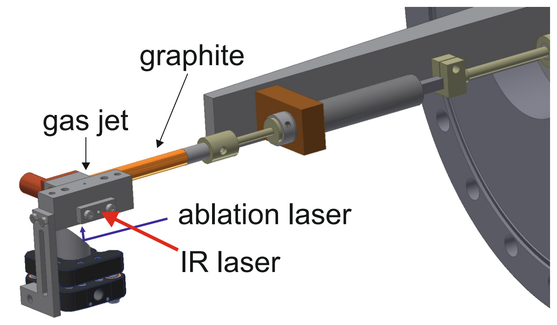
Fast fs beams can be used to ionise samples with lower molecular dissociations than with ns beams, leading to enhanced formation of the molecular ion and orders of magnitude higher efficiencies. Resonant and non-resonant laser ionization can be used to ionise neutral species for selective, sensitive analyses.

- **Time-resolved Angular-resolved Photoemission Spectroscopy TRARPES**



Time-resolved photoemission of hot electrons in dye or quantum dot (QD) sensitised metal oxides. The light absorbing dye is excited with a visible pulse followed by fast transfer of electrons to the oxide conduction band (CB). These hot electrons are then measured using photoelectron spectroscopy with the EUV light. We aim to measure the lifetime of these hot electrons as a function of defect density in the oxide, which is thought to lead to trapping of the electrons and is a major limitation in the efficiency of dye sensitised oxide solar cells. (collaborations with Andrew Thomas, Manchester

- **HHG from (metal) clusters.** Nanoclusters generated with the laser ablation method and characterized with mass spectrometry, IR spectroscopy and quantum chemical simulations.



Acknowledgements

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References

[1]. Shechtman *et al.*, Phase Retrieval with Application to Optical Imaging, IEEE Signal Proc. Mag. **32**, 87-109, 2015.
[2] Poletto *et al.*, Time-delay compensated monochromator for the spectral selection of extreme ultraviolet high-order laser harmonics, Rev. Sci. Instrum. **80**, 123109, 2009.