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The NFFA-SPRINT user facility for ultrafast photoelectron spectroscopy in the 17-31 eV photon energy range based on a coherent narrowband light source up to 200 kHz repetition rate

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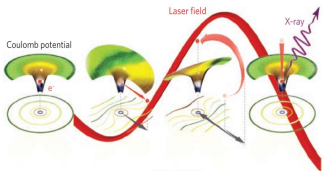
BACKGROUND

A very useful approach in understanding the interplay between charge, spin and orbital degrees of freedom in strongly correlated systems consists in investigating the timescale of the relaxation of the electronic properties after an optical perturbation. However, the prince tool for studying the electronic properties of a material, that is photoemission in all its branches, is difficult to be applied to time-resolved experiments, due to the features of commonly available pulsed sources: infrared or visible light lasers have photon energies too low with respect to the work function of solid samples, whereas Free Electron Lasers (FELs) have too high pulse intensity and too low repetition rate, so that either dramatic space charge effects are produced or a very low electron count rate is achieved.

High-Harmonic Generation (HHG)

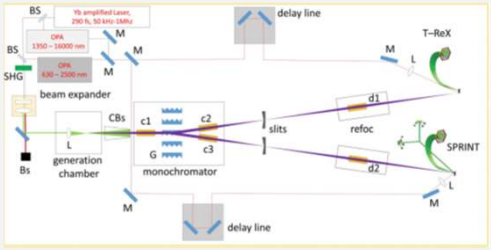
When an **intense monochromatic laser field** is focussed into a **high-pressure gas jet**, the high-order harmonics of the fundamental driving laser are generated. The physics of this process, termed HHG, can be illustrated by a simplified **semiclassical three-step model** [1]:



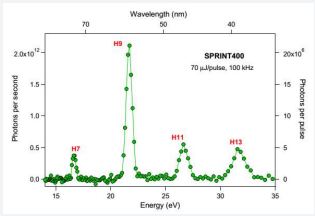
1) **Ionization of the gas atoms** is produced by the strong field of the laser;
2) during an oscillation of the electromagnetic field **the ionized electron absorbs *n* photons;**
3) in the opposite oscillation, a small fraction of the electrons **recombine with the atom** liberating its excess energy as electromagnetic radiation.

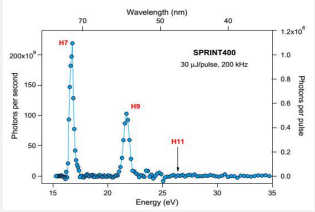
Experimental setup

An experimental set-up has been recently commissioned in Trieste, due to the collaboration of SPRINT-lab (Spin Polarized Research Instrument in the Nanoscale and Time domain) and T-Rex-lab [2]. The system, based on HHG, is able to produce high intensity harmonics **up to the 13th order** of the **driving radiation ($h\nu_0$ = 2.4 eV)**, with tunable repetition rate from single pulse up to 200 kHz. The beamline is equipped with a monochromator specifically designed for ultrafast pulses, whose special geometry allows **to maintain the pulse duration in the scattering process** [3].



The harmonic spectrum, produced with Argon gas, is sensitive to the energy per pulse of the driving laser. The cut-off for **the maximum photon energy depends on the laser intensity *I*** in this way: $h\nu_{max} \approx I/\nu_0^2$.

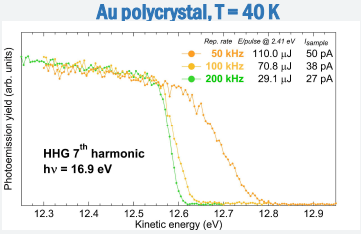


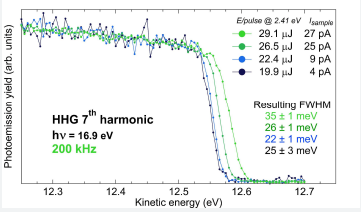


Measurements with calibrated photodiode, and energy selection with 400 lines/mm monochromator.

Space charge mitigation and energy resolution

Au polycrystal, T = 40 K



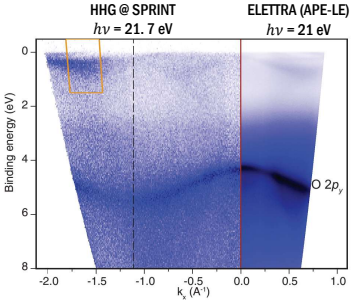


Space charge effects manifest in photoemission spectra as **energy shifts and broadenings**. In our HHG setup, a significant reduction of these effects is obtained **by increasing the repetition rate** (upper figure). A residual space charge shift can be still observed at 200 kHz (lower figure) but the overall energy resolution never exceeds 35 meV. The easy tunability of the intensity of the driving radiation allows to find the good compromise between beam stability and energy resolution in each experiment.

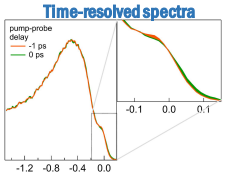
The Fermi edge has been fitted convolving the **Fermi-Dirac function** at fixed T with a **Gaussian distribution** accounting for the experimental energy broadening. For the Gaussian FWHM a lower limit **of 22 meV at photon energy 16.9 eV** has been found.

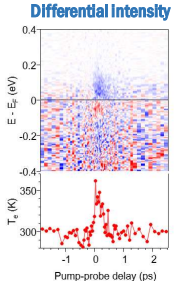
Time-resolved photoemission example

Fe/MgO(001) with O (1x1) reconstruction



The sample is a **40 nm iron film** grown on top of MgO(001) substrate, with oxygen (1x1) reconstruction to prevent contamination [4]. The static **angle-resolved photoemisison (ARPES) cut along Γ-X direction** measured with HHG and synchrotron light are quite similar, with the well recognisable dispersive state originating from O $2p_y$.





Our time-resolved measurement is performed with **pump energy of 1.55 eV and fluence of 0.7 mJ/cm²** (integrating in momentum in the region highlighted in orange in the ARPES image). We observe a broadening of the Fermi edge that can be ascribed to **an increase of the temperature T_e of the thermal electron population**. The non-thermal electrons excited by the pump rapidly (<110 fs, our temporal resolution) transfer their energy to the electron bath via e-e scattering.

The photoemission spectrum $f(E)$ can be modeled as the product of the spectral function $A(E)$, containing the band structure of the material, and the Fermi-Dirac distribution $f_{FD}(E;\mu,T_e)$, convoluted with a Gaussian G accounting for the experimental resolution.

Given the low fluence involved, the spectral function can be considered constant and the transient variation can be attributed only to the Fermi-Dirac distribution, allowing to retrieve the trend of T_e as a function of pump-probe delay, which shows the fast relaxation (<300 fs) almost to the equilibrium value due heat transfer to lattice and spin degrees of freedom.

$$f(E) = [A(E) * f_{FD}(E;\mu,T_e)] \otimes G$$

$$f_{FD}(E;\mu,T_e) = \frac{1}{1+e^{(E-\mu)/kT_e}}$$

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