Opening new doors for photoemission spectroscopy in the VUV

Angle-resolved Photoemission Spectroscopy

In recent years, angle-resolved photoemission spectroscopy (ARPES) has developed into a versatile method for studying the atomic properties of solids. It allows the Fermi edge, the band structure as well as the band gap and the electronic interactions in a sample to be measured directly. Among other topics, ARPES has made important contributions to the understanding of high-temperature superconductivity.

A central component of an ARPES system is the light source initiating the photoelectric effect. Aside from synchrotrons, pulsed laser sources have been used so far. However, their emission is limited to short wavelengths of around 120nm, which also limits the range of elements that can be examined. In order to cover a larger momentum space and, for example, to examine materials such as graphene, higher photon energies, i.e. shorter wavelengths in the vacuum ultraviolet (VUV), are required. Also the pulse duration plays a crucial role in time-resolved ARPES (tr-ARPES).

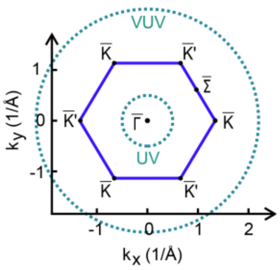

Fig. 1. Brillouin zone of a typical material with high symmetry points. Dashed lines illustrate the increase in observable momentum when using VUV probe pulses compared with typical UV pulses

Both requirements suggest the use of high harmonic generation (HHG) as a VUV light source. In HHG, the nonlinear conversion of femtosecond laser pulses in a gas target yields ultrashort pulses in the extreme ultraviolet or even tender X-ray spectral region [1]. In addition to the HHG stage, a downstream monochromator and focusing stage is required for delivering a monochromatic VUV beam onto the ARPES sample.

Turn-key HHG beamline for VUV-ARPES

The HPS beamLINE VUV is a turn-key VUV light source for ARPES experiments. It consists of a long-term stable ceramic HHG gas target setup that allows for continuous operation for weeks without any maintenance or adjustment. Flux fluctuations <3% over 12 hours ensure reliable extensive measurement campaigns. An elaborate differential pumping configuration increases the photon flux by reducing reabsorption of VUV light by residual gas and ensures UHV-compatibility at the ARPES chamber interface.

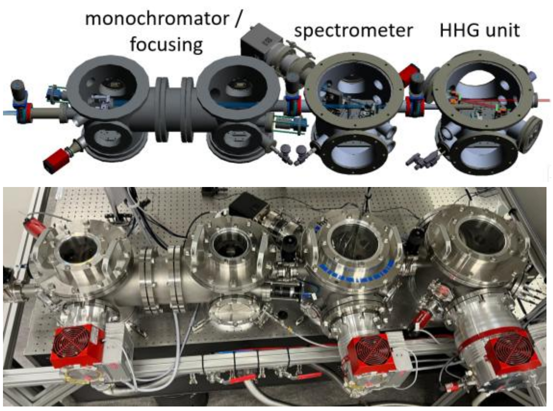

Fig. 2. beamLINE coherent VUV light source

Downstream, a spectrometer provides diagnostics. Via software control it can be inserted into the beampath for a periodic characterization of the flux in the desired harmonic and the suppression of adjacent harmonics.

The subsequent monochromator and focusing stage delivers the VUV beam to the ARPES sample. The focus spot size is 70um, its position can be comfortably controlled in software.

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Incoupling of an auxiliary beam is realized in an optional intermediate stage.

The total system footprint is 1.8 x 0.8m. The focal distance can be set according to experimental requirements.

The frequency-doubled output of a Light Conversion ORPHEUS-OPCPA provides ideal driver pulses for HHG [2]: 400nm-wavelength pulses with 6uJ energy and 20fs duration at 50kHz repetition rate.

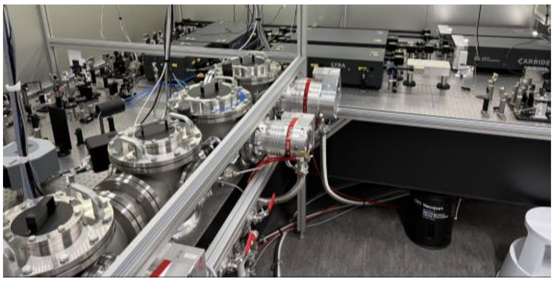

Fig. 3. HPS beamLINE and Light Conversion laser system

The presented beamline was configured for the seventh harmonic at 21.7eV, resp. 57nm. With the Light Conversion laser system, the HPS beamLINE VUV delivers $4 \cdot 10^{12}$ph/s at the source in this harmonic. At the output, $4 \cdot 10^{11}$ph/s are available in the monochromatized harmonic at 21.7eV. This flux level comfortably allows for ARPES and other experiments in the VUV range.

Other harmonics, e.g. at 27.9eV, resp. 44nm can be selected with a minor reconfiguration of the monochromator unit.

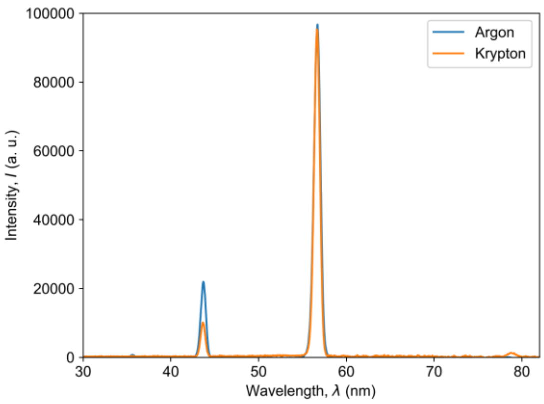

Fig. 4. VUV spectrum of seventh harmonic at 57nm with adjacent harmonics visible (before mono-chromator stage)

With the HPS beamLINE VUV HHG system, an integrated and low-maintenance light source is available for ARPES measurements in the VUV spectral range. The high repetition rate of 50kHz significantly shortens acquisition times over previously available systems operating in the few kilohertz range and opens the door for new studies with higher sensitivity.

References

1. Corkum. Phys. Rev. Lett., 1993, 71, 1994
2. https://lightcon.com/product/orpheus-opcpa/

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