

A compact MHz high-harmonic light source for efficient laboratory photoemission spectroscopy

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Abstract: We demonstrate high-order harmonic generation driven by a compact fiber laser as a light source for efficient photoemission spectroscopy, which allows mapping of the dominant part of the valence band of Ag(100) within 10 seconds.

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1. Introduction

Light sources based on high-order harmonic generation (HHG) have great potential due to their wide photon energy range and the sub-femtosecond pulse duration [1]. Applications of HHG light sources to photoemission spectroscopy and microscopy can therefore provide opportunities for laboratory experiments. Despite the promising properties of the HHG light sources, there are still limitations due to the complicated optical setup and, most importantly for applications in photoemission spectroscopy, due to the relative low repetition rate which is typically in the kHz range. For photoemission spectroscopy and microscopy experiments, the low repetition rate limits the average photoelectron count rate through the space-charge effects coming from the Coulomb repulsion between photoelectrons within one light pulse [2-4].

In this work we present a megahertz HHG setup using a commercial, turn-key ytterbium fiber laser and provide a compact and tunable light source for photoemission spectroscopy. Moreover, by taking advantage of the well-defined pulse structure of the high-order harmonics, we combine the light source with an advanced time-of-flight (ToF) electron spectrometer [5]. This unique combination improves the detection efficiency in photoemission experiments due to the parallel detection of momentum and energy of photoelectrons using state-of-the-art electron optics.

2. Experiment

2.1 High-order harmonic generation

The experimental setup is shown in Fig.1(a). The high-order harmonic generation is driven by a ytterbium fiber laser system with a variable repetition rate from 200 kHz to 25 MHz and a central wavelength at 1040 nm. The laser has a pulse width of 250 fs and an adjustable pulse energy up to 15 μ J. To generate the high-order harmonics, the laser beam is focused by an achromatic lens into an argon or xenon gas jet provided by a glass capillary.

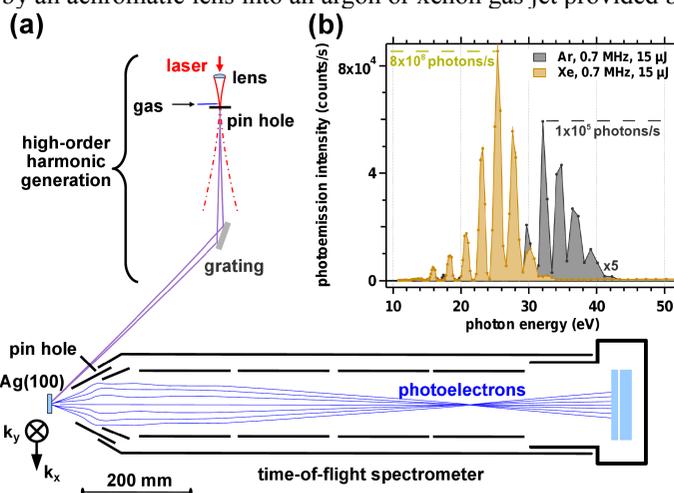


Fig. 1. (a) Setup of photoemission experiments. (b) Spectra of high-order harmonics.

The harmonics are generated in the gas jet and then passed to a toroidal grating, where the harmonics are diffracted and focused on the sample. The harmonic spectrum is measured by turning the grating gradually and using the sample as a mirror to reflect the light on the chevron multichannel plate detector in the ToF spectrometer. As shown in the spectra in Fig1.(b), harmonics with photon energies ranging from 16 to 40 eV are produced and a maximal flux of 10^9 photons/s at 25 eV is available at the sample position.

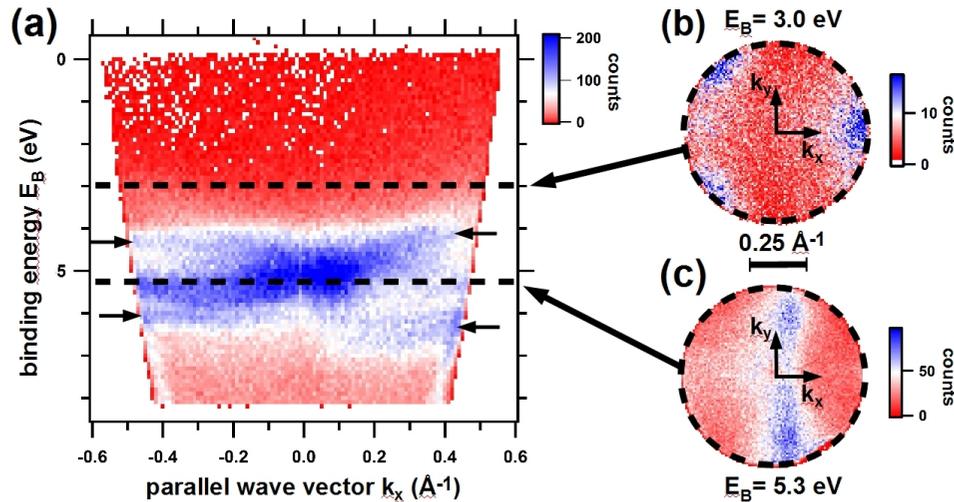


Fig. 2. Photoelectron distribution from Ag(100) using the 19th harmonic generated in xenon. The photon energy is 22.7 eV and the light is *p*-polarized.

2.2 Efficient photoemission spectroscopy using the high-order harmonics

We use the 19th harmonics (22.7 eV) which delivers 10^8 photons/s of *p*-polarized light at the sample position and at the chosen repetition rate of 1MHz. Photoelectrons emitted within ± 15 degrees around the surface normal are collected simultaneously by an electrostatic ToF spectrometer, which is mounted at 45 degrees with respect to the incident light. The efficiency of the setup is demonstrated in photoemission experiments on Ag(001) as shown in Fig.2. The momentum-resolved valence band data from Ag(100) shown in Fig.2(a) have been measured within a total data acquisition time of 10s and show dominant features with flat dispersion from the Ag *d* bands.

The photoemission intensity exhibits a clear asymmetry between positive and negative k_x sides as indicated by the arrows in Fig.2(a). This asymmetry is also clearly visible in an uneven momentum distribution displayed in Fig.2(b,c). The Ag(001) surface has a four-fold symmetry and the linear polarization of light is 45 degrees tilted from the surface normal, which has only an in-plane component parallel to the k_x momentum direction. Therefore the incident light breaks the original four-fold symmetry and leads to a two-fold photoemission pattern with only a mirror-symmetry about the k_x axis.

3. Summary

A compact high-order harmonic light source with megahertz repetition rates has been developed and it is applied for photoemission spectroscopy in combination with time-of-flight (ToF) spectrometer. We demonstrate efficient photoemission experiments which are capable of mapping the momentum-resolved valence band structure within 10 seconds without degradation from space-charge effects.

4. References

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