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Oscillator-based High-order Harmonic Generation at 4 MHz for Applications in Time-of-Flight Photoemission Spectroscopy

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Abstract. We generate high-order harmonics at 4 MHz by a long-cavity laser and demonstrate applications in time-of-flight photoemission spectroscopy. Our results suggest a straightforward, oscillator-only setup for HHG-based photoelectron spectroscopy and microscopy with high repetition rate.

1 Introduction

High-order harmonic generation (HHG) allows to produce attosecond pulses of coherent vacuum-ultraviolet light and x-ray using a laboratory setup. The applications of HHG-based light sources in photoelectron spectroscopy and microscopy have great potential for studies of ultrafast electron dynamics in atoms, molecules as well as condensed matters. Although HHG was discovered more than two decades, it is still at the frontier of research to develop HHG setups with a high repetition rate beyond megahertz frequency.

Conventional HHG experiments are driven by amplified laser systems at a repetition rate of several kilohertz [1–3]. As a consequence, the statistics of measurements are limited to a few thousand events per second and a long acquisition time is required for applications in photoelectron-based spectroscopy and microscopy [4–7] (Fig. 1). To significantly increase the repetition rate of HHG, three conceptually different approaches were demonstrated recently. By using high power fiber-based lasers and amplifiers, Vernaleken et al. generate harmonics up to the 17th order at a repetition rate of 20.8 MHz [8]. In their setup, the generation of sufficient power to drive the harmonics and the compression of high power laser pulses are demanding. As an alternative, Kim et al. take advantage of the actual nanotechnology to fabricate optically triggered plasmonic antenna, aiming at the resonantly enhanced near-field to drive HHG at 80 MHz [9]. However, due to the damages caused by the intense electric field, their results can not be reproduced by other groups and the ideas are currently under critical debate [10]. The desired enhancement of the laser electric field at high repetition rate, nevertheless, can be achieved in a macroscopic resonant cavity, either external to or integrated inside the driving laser [11, 12]. With enormous efforts optimizing the output coupler for harmonics, the repetition rate of HHG using an external cavity goes nowadays beyond 150 MHz [13].

In this contribution, we report a more straightforward design for HHG at megahertz repetition rate. In our setup, we use directly the output of a Ti-sapphire laser at 4 MHz and focus it into a gas jet with a high backing pressure. Furthermore, we apply the HHG setup as a light source for time-of-flight photoemission spectroscopy. Our results suggest a compact setup for HHG at megahertz repetition rate and provide the basis for applications in photoelectron spectroscopy and microscopy.

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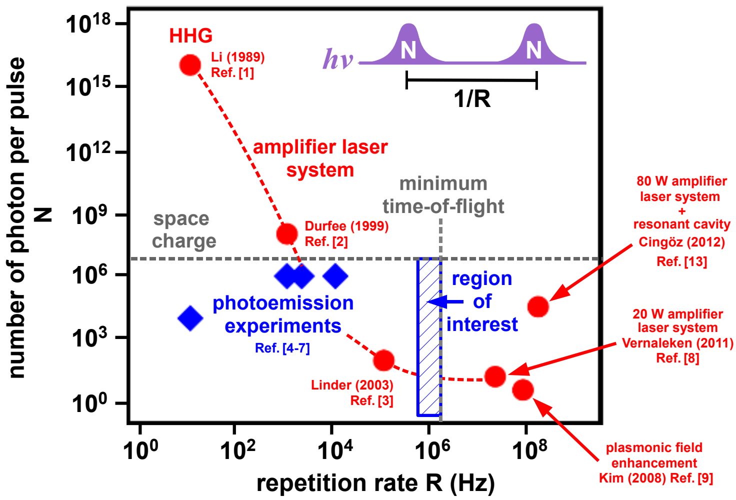

Fig. 1. Overview of the pulse structure of HHG setups (red circles, [1–3, 8, 9, 13]) and HHG-based light sources for photoelectron spectroscopy at surfaces (blue diamond, [4–7]). Dashed lines indicate the limits due to Coulomb repulsion between photoelectrons (space-charge effect) and the finite time-of-flight of photoelectrons. The marked region-of-interest can provide optimal condition for time-of-flight photoelectron spectroscopy and microscopy.

2 Experiment

2.1 Characterization of high-order harmonics

The harmonic generation is driven by a commercial long-cavity Ti-sapphire laser (Femtosource scientific XL650). The laser has a repetition rate of 4 MHz and the central wavelength is at 800 nm. The laser pulse has a width of 50 fs and a pulse energy of 650 nJ. To generate high-order harmonics, the laser beam is expanded to a diameter of 15 mm and then focused into a gas jet in a generation chamber. The experimental setup is shown in Fig. 2(a). The achromatic focus lens has a focal length of 50 mm and the laser focus has a diameter of 5 μm. The gas jet of argon or xenon is provided by a borosilicate glass capillary with an opening of 30 μm and a high backing pressure up to 3.5 bar. The harmonics are generated in the gas jet and then passed to a monochromator chamber through a 200 μm pin hole for differential pumping. In the monochromator chamber, the harmonics are diffracted by a toroidal grating and focused on the channelplate detector coupled to a phosphor screen. The fluorescence from the phosphor screen is measured by a CCD camera, which can be operated in an event-counting mode for determination of the count rate. The HHG spectrum generated by an argon jet with a backing pressure of 3.5 bar is presented in Fig. 2(b) with estimated count rates.

2.2 Time-of-flight photoemission spectroscopy using high-order harmonics

We use the generated harmonics as an excitation source for time-of-flight photoemission spectroscopy. The channelplate detector in Fig. 2(a) is replaced by a Cu(111) sample located in an ultrahigh vacuum chamber. In addition, a pin hole with a diameter of 1.5 mm is placed after the monochromator to filter the chosen harmonic as well as to assist differential pumping. Photoelectrons are collected by an electrostatic time-of-flight spectrometer (Themis 1000, SPECS Surface Nano Analysis GmbH), which is mounted at 45 degrees with respect to the incident light. The sample is positioned in normal emission geometry. In Fig. 2(c) the valence band photoemission data for a photon energy of 14 eV (9th harmonic from a Xe jet) is shown. All photoelectrons with photoemission angles between ±15 degrees were recorded simultaneously. Near the Fermi level (E_F), we observed the characteristic parabolic dispersion of the Shockley surface state (blue dashed curve). At lower energies around 2 eV below E_F, we observed less dispersive features which are attributed to the copper d-bands.

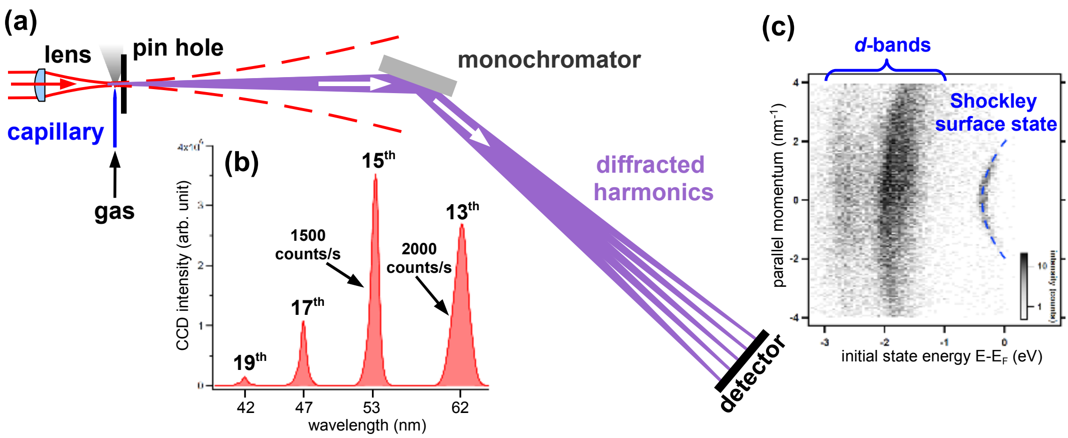

Fig. 2. (a) Setup for generation and characterization of high-order harmonics. (b) The spectrum of harmonics generated from argon with a backing pressure of 3.5 bar. (c) Photoemission spectra from Cu(111) along the $\overline{\Gamma K}$ direction using the 9th harmonic generated in xenon. The photon energy is 14 eV and the light is *p*-polarized.

3 Summary

We demonstrate a compact design of high-order harmonic generation at 4 MHz repetition rate using the direct output of a femtosecond laser cavity. Moreover, we use the harmonics as a light source for time-of-flight photoemission spectroscopy. Our results suggest a straightforward, easy to operate and affordable method to generate high-order harmonics and to provide a basis for HHG-based photoelectron spectroscopy and microscopy with excellent statistics.

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