High-order harmonic light source at megahertz for double photoemission spectroscopy of correlated electrons

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Abstract: We develop high-order harmonic generation at repetition rates up to 1 MHz using an ytterbium-fiber laser and demonstrate the first laboratory double photoemission experiments with high-order harmonics to study strongly correlated materials.
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1. Introduction

High-order harmonic generation (HHG) from atoms driven by intense laser systems can produce radiation from vacuum-ultraviolet up to the hard x-ray range. The application of HHG light source for spectroscopy and microscopy experiments has great potential due to the possible time resolution in a pump-probe experiment. However, for photoelectron spectroscopy experiments there is a technical limitation due to space-charge effects which originate from the Coulomb repulsion between photoelectrons generated by a single light pulse. This limitation can be circumvented by increasing the repetition rate of HHG up to megahertz range, which is several orders of magnitude higher beyond the low repetition rate of traditional Ti-sapphire laser amplifier system at kilohertz.

On one hand, the HHG light source at high repetition rate can significantly boost the efficiency of future attosecond time-resolved photoemission spectroscopy and microscopy since the maximum photoelectron intensity without degradation of space-charge effects scales linearly with the repetition rate of the light source. On the other hand, the high repetition rate of the light source allows applications for advanced spectroscopic methods, such as the double photoemission (DPE) experiments, which require otherwise synchrotron radiation or a much longer acquisition time for light sources at lower repetition rate. In DPE experiments two photoelectrons are excited by a single light pulse. Both electrons are analyzed with energy and momentum resolution and the correlation between both are extracted directly.

In this contribution we demonstrate high-order harmonic generation at megahertz repetition rate and use it as a light source for double photoemission spectroscopy experiments. We show photon energy dependent measurements ranging from 18 to 42 eV for Ag(100), which demonstrate the capability of our light source in combination with a state-of-the-art time-of-flight photoemission spectrometer. The combination with two time-of-flight spectrometers is used for double photoemission experiments on the strongly correlated material NiO. Our work shows a unique application of high-order harmonic light source for laboratory photoemission spectroscopy, which is suitable for general material investigations as well as for specific advanced DPE spectroscopy on strongly correlated materials [1].

2. Experiments

2.1 Photoemission experiments with tunable photon energy using the high-order harmonics

In Fig.1(a) the experimental setup is shown. The high-order harmonics are generated by focusing the output of the fiber laser into a 30 μm argon gas jet. About 5 mm after the laser focus the fundamental beam is blocked by a 200 μm pin hole, whereas the produced harmonics propagate through the pin hole [2]. Then the harmonics are diffracted by a toroidal grating and focused on the sample. The spectrum of the harmonics is measured by reflection from a Ag(100) surface to a multichannel plate detector and is shown in Fig.1(b). Photon energies ranging from 20 to 40 eV are generated with a maximum photon flux of about $1x10^5$ photons/s at 32 eV, as estimated by the reflectivity of the silver surface and an assumed 10% detection efficiency of the detector.

To demonstrate the unique capability of the harmonic light source for photoemission experiments we perform photon energy dependent measurements of the momentum-resolved Ag(100) valence band structure. Photoelectrons emitted from the sample within ±15 degrees are collected and analyzed energy- and momentum-resolved. The energy-momentum distribution of photoelectrons for selected photon energies are shown in Fig.1(c). Here we observed characteristic features from the Ag *d* bands at binding energies between 4 and 8 eV. The dispersive features at lower binding energies and with weaker intensity are identified as the *sp* bands.

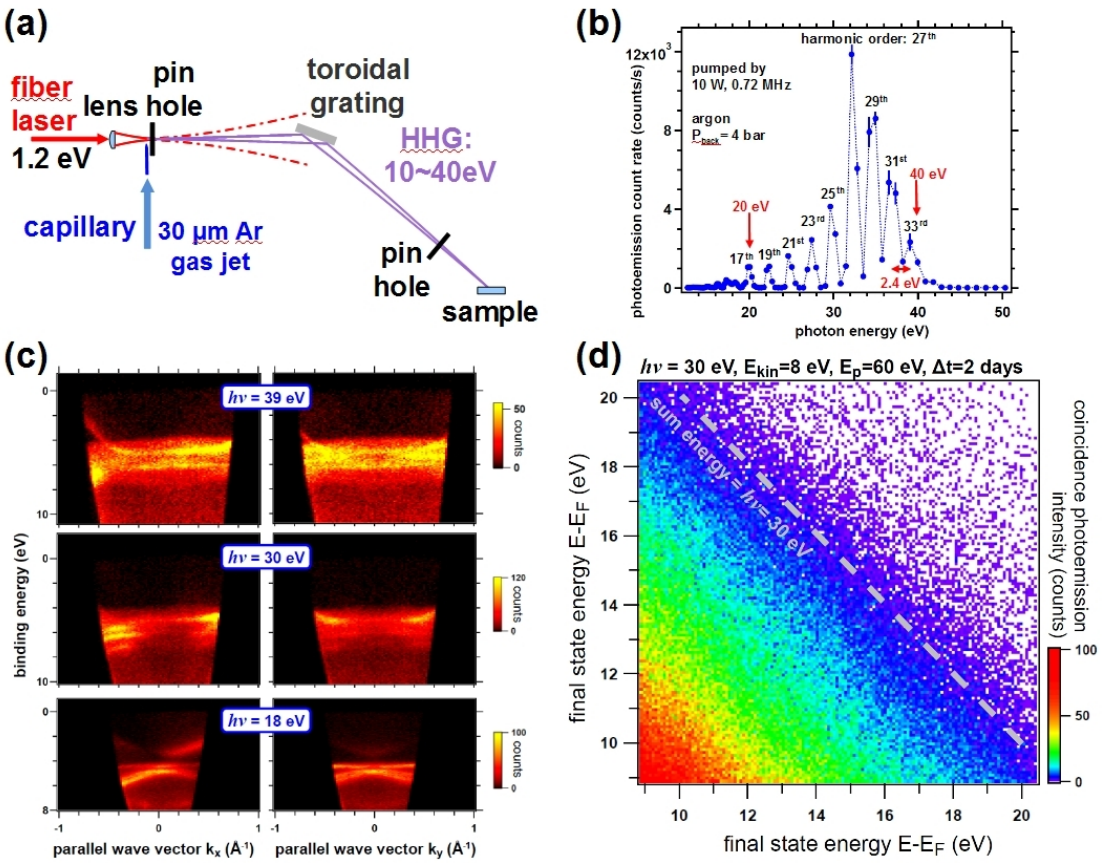

Fig. 1. (a) Setup for high-harmonic generation. The output of the fiber laser is focused into a gas jet and the generated harmonics are selected and focused onto the sample by a toroidal grating. (b) The spectrum of generated harmonics ranging from 20 to 40 eV. (c) Photon energy dependent photoemission spectra measured on Ag(100) surface showing characteristic dispersion of the *sp* bands from 0 to 4 eV and the *d* bands from 4 to 8 eV. (d) Double photoemission spectra measured on NiO films. The horizontal and vertical axes indicate the energies of the first and the second photoelectron when detected in coincidence. The diagonal dashed line marks the cutoff expected if both electrons share the maximal available energy. The acquisition time for this measurement is two days.

2.2 Double photoemission experiments on NiO films

We demonstrate proof-of-principle double photoemission (DPE) spectroscopy on NiO films to investigate the signature of electron correlation in this material. In the experiments two photoelectrons are detected within a time window of 100 ns. In Fig.1(d) the energy distribution of photoelectron pairs is displayed with two energy axes, which indicate the energy of the two individual electrons in each pair. For correlated electrons we expect an energy sharing between both electrons. Consequently the maximum energy which is approximately given by the photon energy minus twice the work function defines the upper limit for correlated electron pairs. This limit is marked as dashed line in Fig.1(d) and is clearly visible in the coincidence data. The DPE intensity increases as the sum of the energies of individual electrons in a pair is lower. At a given sum energy, the coincidence spectra show rather flat dependencies on the energy difference between electrons in the pairs.

4. Summary

We have presented a high-order harmonic generation (HHG) light source with megahertz repetition rate aiming at application for double photoemission spectroscopy experiments. The light source allows photoemission experiments with tunable photon energies from 20 to 40 eV. As a proof-of-principle experiment we performed the first laboratory double photoemission spectroscopy using a HHG light source to study the two-dimensional energy distribution of photoelectron pairs for strongly correlated NiO films.

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