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Photoelectron spectroscopy using high-order harmonics at megahertz repetition rates: study of correlated electron pairs in solids

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With the progressive development of femtosecond lasers, high-order harmonic generation (HHG) has become a standard technique to produce a widely tunable photon energy spectrum in the laboratory. However, the application of HHG-based light sources to electron spectroscopy and microscopy has been limited by the space-charge effects at kilohertz repetition rates of high power laser systems. With our recent development of an HHG-based light source at megahertz repetition rates using a compact fiber laser, we have demonstrated an efficient combination with time-of-flight electron spectrometer for laboratory photoelectron spectroscopy [1,2].

In this contribution we will report the advanced application of our MHz HHG-based light source to double photoemission (DPE) spectroscopy on solids, where pairs of correlated valence electrons are photoexcited and analyzed with energy- and momentum-resolution. With photon energies of 25 and 32 eV, we discover detailed structures in the energy distribution of electron pairs from Ag(001) and Cu(111) [3]. In the two-dimensional DPE energy spectra, distinct features with well-defined sum-energies of electron pairs are identified. These two-electron features can be attributed to the emission of correlated sp- and d-electrons as well as to two correlated d-electrons. The results will be compared with the self-convolution of density of states and discussed in terms of electron correlation.

References:

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