Core-level Spectroscopy at FLASH and Investigation of Space Charge Effects

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The Free–Electron Laser in Hamburg (FLASH) produces highly brilliant, ultrashort, and coherent pulses in the VUV regime. This offers the possibility for time–resolved pump–probe photoemission experiments as well as to study the photoemission process at extreme photon densities. The most relevant limiting factor for these experiments will be the so–called space-charge effect (SCE) (see figure 2(B)), which occurs when a huge amount of electrons is leaving the sample in a short timescale. The electrons then influence each other by Coulomb forces in a way that the original photoemission spectra becomes blurred and shifted. SCEs in photoemission experiments have already been observed and studied with 3^{rd} generation synchrotron radiation sources [1] as well as with high intensity femtosecond lasers [2]. While energy shifts and broadenings in both experiments are of the order of 10 to 100 meV, SCEs of up to some eV are expected and have already been observed in earlier experiments [3] using FEL radiation.

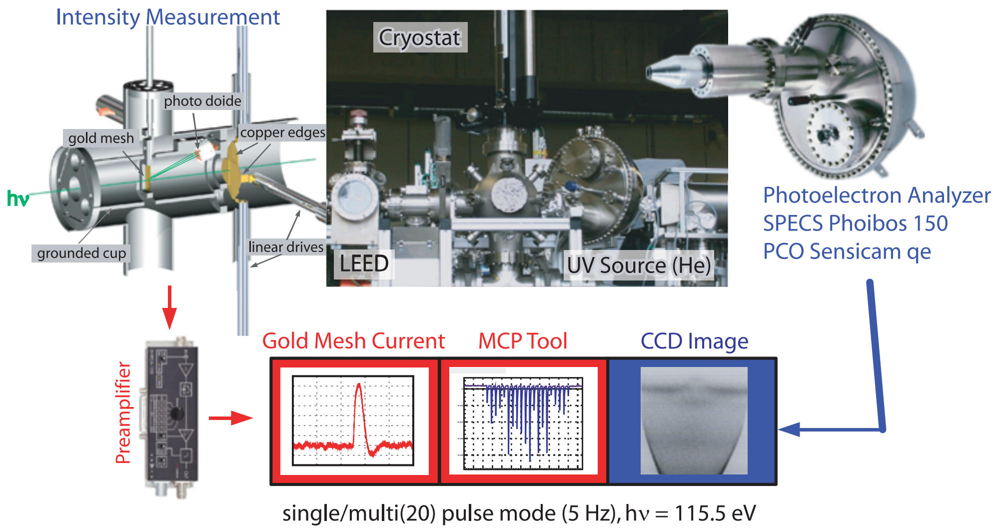

Figure 1: Experimental setup for photoelectron spectroscopy at FLASH beamline PG2 with main components: photoelectron analyzer with 2D–CCD detector, Cryostat, LEED, home built pulse–intensity measurement tool with preamplifier. Photon pulse intensities are measured by gold mesh current as well as with an MCP based tool.

Employing highly intense monochromatic VUV-pulses ($h\nu$ = 115.5 eV, FEL 3^{rd} harmonic) delivered by the PG2 beamline of FLASH [4] we performed angle resolved as well as core–level photoelectron spectroscopy on the transition metal dichalcogenide $1T$–TaS_2 in the charge density wave (CDW) phase (T = 140 K). FLASH was operated in the single as well as in the multi pulse (20) mode with a repetition rate of 5 Hz. One characteristic of FEL radiation is the fluctuating intensity from pulse to pulse. Hence, it was essential to store the spectra and the photon intensities for every single pulse train. Afterwards the spectra had to be sorted according to the photon intensity and summed up to obtain decent statistics. Photoemission spectra were taken with a SPECS PHOIBOS 150 analyzer employing a PCO Sensicam qe camera. The pulse intensities were taken with a home built tool, measuring a gold mesh current with a LeCroy oscilloscope after preamplification with a Femto DHPCA-100 ultrafast preamplifier. The other method for pulse intensity measurements was a MCP- based tool positioned between the monochromator and the experiment (see figure 1).

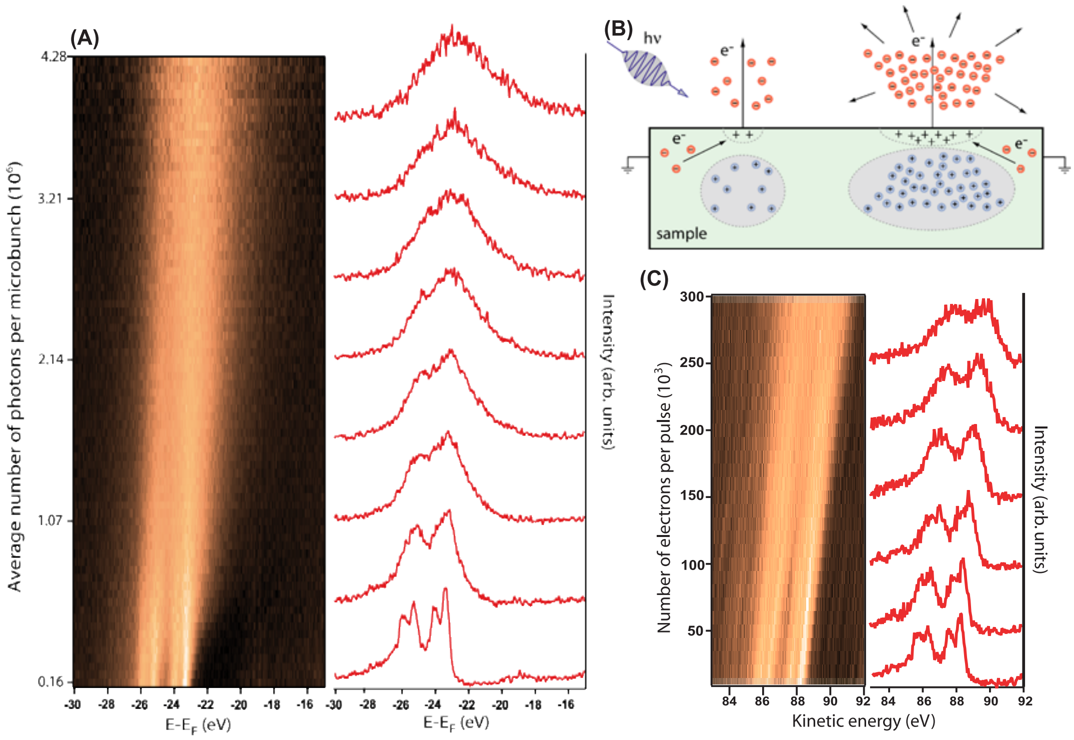

Figure 2: (A) Ta 4f core-level spectra of 1T–TaS_2 in the CDW phase (T = 140 K), taken at hν = 115.5 eV in the multi bunch mode and sorted according to the average number of photons per microbunch. (B) Schematic drawing of SCE: Excitation of electrons with pulses of different intensity leaving positive mirror charges in the sample. (C) First simulations of SCEs on the Ta 4f core-level spectra using the Treecode Algorithm.

In Fig. 2(A) we show pulse intensity sorted Ta 4f core-level spectra. In the bottom spectrum corresponding to the lowest pulse intensity we can still observe the typical four-peak structure of the Ta 4f core–level in CDW phase. With increasing photon numbers and thus increasing number of photoelectrons per pulse this structure becomes blurred as well as shifted towards higher kinetic energies. To understand the observed SCEs, we have performed self–consistent N–body simulations based on the Barnes & Hut Treecode Algorithm [5] originally developed for simulating planetary movements (see fig. 2(C)). The observed trends with regard to energy broadening and shift are in good agreement with the experiment. The comparison between the experimental results and the results of the simulations suggests that photoemission is possible up to $4 \cdot 10^5$ photons per 100-fs-pulse, corresponding to about $4 \cdot 10^4$ photoelectrons.
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