Contribution ID: **187** Type: **Contributed talk**

New Insights Into the Laser-Assisted Photoelectric Effect from Solid-State Surfaces

Thursday 29 August 2024 18:00 (15 minutes)

Recent advances in high-intensity ultrafast X-ray sources enable a new era of time-resolved pump-probe experiments, revealing hitherto inaccessible information about the interactions of photons with surfaces and the electronic dynamics they induce. Femtosecond time-resolved laser-assisted photoemission (tr-LAPE) provides a deeper understanding of the very foundations of the surface photoemission process. When X-ray and optical laser pulses overlap in time and space, the photo-ejected electron is accelerated by the intense optical laser field, reflecting the dynamics of the laser-surface interaction. This leads to the creation of sidebands in the photoelectron spectrum corresponding to the absorption and stimulated emission of photons in the laser field.

We present a systematic femtosecond time-resolved investigation of the LAPE effect in two similar metallic solids - W(110) and Pt(111) single crystals - investigated by ultrafast X-ray photoemission spectroscopy at the soft X-ray free-electron laser FLASH at DESY in Hamburg (cf. Fig. 1). Surprisingly pronounced differences in the LAPE spectra of the two materials measured under identical conditions are observed. While tungsten exhibits strong sidebands up to the sixth order, sidebands in platinum are barely discernable beyond the second order. The unexpected observations are explained on a semi-quantitative level by analyzing the dynamic dielectric responses of the two materials, demonstrating new insight into the time-dependent IR field strength in the surface region of the metals, and the dynamics of the light-matter interaction. Calculations using the strong-field approximation show that an intrinsic, near-surface IR laser field enhancement by a factor of 4 is the root cause for the larger number of sidebands in tungsten compared to platinum.

These new insights demonstrate the exceptional sensitivity of femtosecond tr-LAPE measurements to the dielectric properties of solids in a sub-nm thick surface layer. The findings may inspire new techniques to monitor electronic and lattice dynamics in surface regions and help to gain a deeper understanding of surface electromagnetic fields including local fields near chemisorbed atoms and molecules.

Moreover, a detailed understanding of LAPE contributions in high-resolution pump-probe photoemission spectroscopy is a prerequisite for correctly interpreting a growing number of pump-probe experiments at next-generation, high repetition-rate light sources. Therefore, the new results provide new opportunities for even more challenging experimental and theoretical investigations utilizing ultrafast and intense laser fields.

Figure caption:
Fig. 1: Femtosecond time-resolved XPS spectra of the (A) W 4f and (B) Pt 4f photolines as a function of time-delay (vertical) and binding energy (horizontal).

I plan to submit also conference proceedings

No

Primary authors: WENTHAUS, Lukas (FS-FLASH-O (FLASH Scientific User Operation)); Dr KABACHNIK, Nikolay (European XFEL GmbH); Dr BORGWARDT, Mario (Lawrence Berkeley National Laboratory); PALUTKE, Steffen (Eur.XFEL (European XFEL)); KUTNYAKHOV, Dmytro (DESY Photon Science); PRESSACCO, Federico (FS-LA (Laser Operations & User Support)); SCHOLZ, Markus (FS-FLASH (FLASH)); POTOROCHIN,

Dmitrii (FS-PETRA-S (FS-PET-S Fachgruppe P04)); WIND, Nils (FS-SXQM (Soft X-ray Spectroscopy of Quantum Mat.)); DUESTERER, Stefan (FS-FLASH-D (FLASH Photon Diagnostics and Controls)); BRENNER, Guenter (FS-FLASH-B (FLASH Photon Beamlines and Optics)); Dr GESSNER, Oliver (Lawrence Berkeley National Laboratory); MOLODTSOV, Serguei (Eur.XFEL (European XFEL)); EBERHARDT, Wolfgang (DESY); ROTH, Friedrich (XFEL_EET (Soft X-Ray Port))

Presenter: ROTH, Friedrich (XFEL_EET (Soft X-Ray Port))

Session Classification: Mikrosymposium 12/2: Time Resolved Techniques

Track Classification: 12. Time resolved techniques