Ultrafast electron dynamics at surfaces studied using femtosecond photoelectron spectroscopy techniques

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Synopsis: We directly observed the core-level relaxation dynamics in a complex surface-adsorbate system by combining laser-assisted Auger decay (LAAD) with the laser-assisted photoelectric effect (LAPE). Furthermore, we show that by using longer-wavelength driven pulses, we can more-effectively probe ultrafast dynamics using LAPE. These findings show how complex electron dynamics in solid and surface systems can be observed on femtosecond-to-attosecond time-scales.

Atomic and molecular dynamics at surfaces is a topic of importance to many processes of both practical and fundamental interest, including catalytic chemical reactions, detector photocathodes, and fundamental explorations of image potential states on surfaces. In many of these processes, the relevant time scales span from picoseconds to attoseconds. The ability to generate extremely short-duration light pulses using high harmonic generation now makes it possible to observe even the fastest surface dynamics. However, the development of new experimental methods has also played a key role in this area. In particular, the use of the laser-assisted photoemission (LAPE) and laser-assisted Auger decay (LAAD) processes has made it possible to study high-energy electronic processes where the dynamics are very fast.

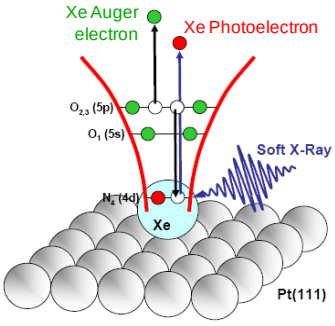

Fig. 1. Schematic of the measurement of the core-level lifetime in atoms on a surface

In previous work, we reported the first observation of LAPE in solids. More recently, we presented the first direct time-resolved observations of the lifetime of core-excited states of an atom adsorbed on a surface. By comparing the LAPE signal from direct photoionization of the substrate with the delayed LAAD signal from Auger decay of the adsorbate, we measured the lifetime of the $4d^{-1}$ core level of xenon on Pt(111) to be 7.1±1.1fs. This result opens up a new area of time-domain surface dynamics where other measurements may provide incomplete information.

Using ultrashort Ti:sapphire laser pulses at $\lambda \approx$ 780 nm, an intensity of $\approx 10^{11}$ W/cm^2 is required to generate an observable LAPE signal. At such high intensities, other effects, such as above-threshold ionization (ATI) and space charge, can influence the spectrum strongly, making it harder to detect a LAPE signal. We show that by driving LAPE with longer-wavelength pulses, the amplitude of the LAPE signal can be significantly enhanced while ATI and space charge effects are suppressed. This finding will make possible ultrafast studies of surface-adsorbate systems and attosecond electron dynamics over a wide range of parameters, for example at low electron energy, or for adsorbates that would otherwise be desorbed or dissociated.

References

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