Towards the Absolute Timing of Photoemission from Condensed Matter Systems

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Abstract: We introduce a viable scheme for measuring the absolute duration of photoemission from solids. It employs an atomic chronograph on the surface during attosecond streaking spectroscopy. First experimental results on a tungsten(110) surface are presented.

OCIS codes: 320.7120, 240.6648.

1. Motivation

The advent of attosecond light sources ($1\text{as} = 10^{-18}\text{s}$) allows the observation of electron dynamics on their natural timescale and therefore the examination of some of the most fundamental processes in physics. In attosecond streaking spectroscopy, photoelectrons are emitted from a gas or solid by an extreme ultraviolet (XUV) light pulse of sub-femtosecond duration. Their momentum is shifted proportionally to the vector potential of an additionally incident near infrared (NIR) light pulse at the instant of photoemission (PE) [1]. This allows to measure relative PE delays between different electronic states [2, 3]. The first attosecond streaking experiment on condensed matter showed that PE from the tungsten 4f core levels in a W(110) surface is delayed by $\Delta\tau_{W4f/WCB} = (110 \pm 70)$as with respect to PE from the conduction band [2]. In contrast, absolute timing of PE remains challenging. Here, we present an experimental approach and first results on the absolute timing of PE from solids.

2. Concept

Measuring the absolute PE timing at a surface requires the definition of a time-zero, i.e., the exact moment that the XUV pulse reaches the surface. Our approach is to adsorb iodine atoms at low coverage on a W(110) surface. PE from this adsorbate layer will happen at a constant time after the absorption of an XUV photon, $\tau^{\text{atomic}}_{\text{orbital}}$, which is intrinsic to the atom and the orbital the photoelectrons emerge from [3]. The adsorbed iodine atoms can therefore be used as atomic chronograph giving access to time-zero, as long as a $\tau^{\text{atomic}}_{\text{orbital}}$ is known. To eliminate residual transport effects, i.e., due to NIR field screening within the iodine overlayer, we performed a coverage dependent study. Extrapolation to zero coverage then allows to extract an undisturbed PE delay. By using iodine, we can take advantage of the large I4d photoionization cross section in the XUV spectral range, which permits accurate streaking measurements, even for sub-monolayer coverages. Iodine forms a saturated atomic monolayer on tungsten at room temperature [5], simplifying the calibration of static XUV spectroscopy for coverage determination. Coverage was tuned via partial thermal desorption. Saturated monolayer as well as sub-monolayers were found to be stable under NIR-intensities needed for streaking spectroscopy.

Absolute PE timing of helium 1s electrons, $\tau^{\text{atomic}}_{\text{He1s}}$, can be determined via ab inito simulations [4]. Hypothetically, this allows access to $\tau^{\text{atomic}}_{\text{I4d}}$ by measuring the PE delay of I4d with respect to He1s in a gas-phase streaking experiment.

3. Results and Discussion

The used experimental setup is described in [6, 7]. The XUV pulses have a central energy of ~105eV with a spectral bandwidth of ~4.9eV. Combining streaked PE spectra at different relative XUV-NIR-Delays yields a streaking

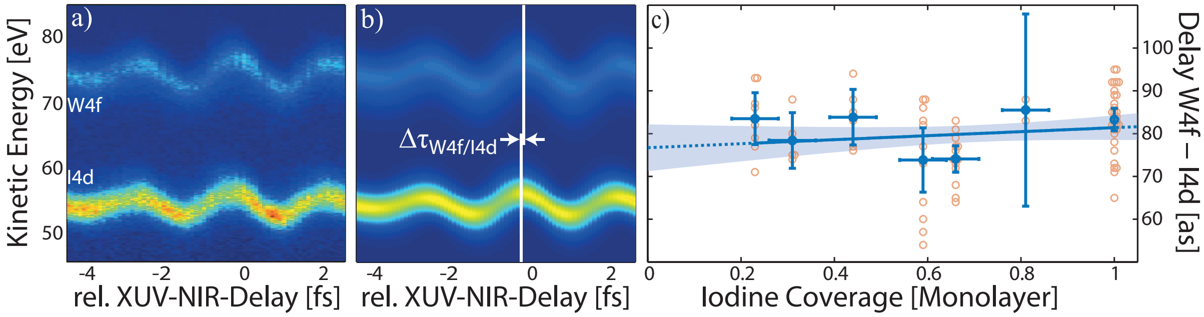

Fig. 1. a) Photoelectron streaking-spectrogram of 0.66 ± 0.05 monolayers of iodine on W(110) as a function of time delay between XUV and NIR pulses. Time step size is 100as. b) Retrieved spectrogram c) W4f delay with respect to I4d, $\Delta\tau_{\mathrm{W4f/I4d}}$, versus iodine coverages. (Red: Individual Measurements. Blue dots: Means over equal coverage. Blue curve: Linear fit. Delay error bars and shaded area represent 95% confidence intervals, calculated assuming a Student's t-distribution. Coverage error is conservatively estimated to 0.05 monolayers.)

spectrogram as depicted in Fig. 1 a). Relative time delays between PE from W4f and I4d orbitals, $\Delta\tau_{\mathrm{W4f/I4d}}$, are extracted by fitting an analytic solution of the time-dependent-Schrödinger-equation (Fig. 1 b)) [3, 7]. A preliminary result of 79 individual streaking spectrograms is shown in Fig. 1 c). To quantify residual transport effects within the iodine overlayer, we performed streaking measurements for 7 different coverages, ranging from a saturated monolayer down to 0.23 ± 0.05 monolayers. The data suggests a coverage dependence of the time delay $\Delta\tau_{W4f/I4d} = (77 \pm 6)\mathrm{as} + \theta(5 \pm 8)\mathrm{as}$, where θ is the iodine surface coverage in units of monolayers. A positive delay means electrons from the I4d orbitals are emitted earlier. The effect of the iodine layer on the delay appears to be on the order of the error of the layer-thickness-independent component of the delay. This justifies the extrapolation towards zero iodine coverage to eliminate effects of the overlayer. The preliminary absolute PE time from the 4f orbitals of a W(110) surface at 105eV excitation energy is stated in Eq. (1). The measurement of $\tau_{\mathrm{I4d}}^{\mathrm{atomic}}$ is currently prepared.

$$\tau_{\mathrm{W4f}} = \Delta\tau_{\mathrm{W4f/I4d}} + \tau_{\mathrm{I4d}}^{\mathrm{atomic}} = (77 \pm 6)\mathrm{as} + \tau_{\mathrm{I4d}}^{\mathrm{atomic}} \tag{1}$$

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