Accessing Energy-Dependent Photoemission Delays in Solids

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Abstract: Our new detection scheme combines the RABBITT technique in solids with simultaneous measurements in a reference argon target. The experiment resolved attosecond delays in the photoemission from noble metal surfaces beyond simple ballistic transport.

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1. Introduction

Studying attosecond delays involved in photoemission is of great interest for our understanding of fundamental processes in nature. Recent developments in the generation of extreme-ultraviolet (XUV) attosecond pulses (1 as = 1×10^{-18} s) provide powerful tools to access such dynamics that unfold on atomic time scales [1]. So far, two main techniques were employed to study photoemission delays in atoms: attosecond streaking [2] and RABBITT [3]. In the case of streaking, a single attosecond pulse is used to ionize the target in the presence of a delayed infrared (IR) reference pulse, while for RABBITT the excitation occurs via a train of attosecond pulses. In both cases information about the photoemission dynamics can be extracted from the delay dependence of the photoelectron spectrum [4,5]. Despite the good results obtained with gaseous samples, the extension of attosecond techniques to condensed matter is not trivial. Phenomena absent in rare-gas samples like space charge, secondary electrons and above threshold photoemission (ATP) by the IR field, limited the applicability of existing methods to high excitation energies (~90 eV). Indeed, so far, only attosecond streaking has been successfully employed in time-resolved studies of photoemission from solids. In 2007 Cavalieri *et al.* [6] studied the delay between the electrons emitted from the *4f* and conduction band in W(110) and found it to be 110 as. Five years later the same group showed that no delay exists in the relative photoemission between the *2p* and valence band of Mg(0001) [7].

We present the first extension of the RABBITT technique to solid-state samples. Compared to streaking, RABBITT exhibits several advantages: (*i*) the required IR intensity is lower (~1×10^{11} W/cm^2), thereby enabling its application even at lower XUV photon energies; (*ii*) the XUV generation scheme is simplified since no single attosecond pulse is required and the carrier-envelope phase of the driving pulse does not have to be stabilized; (*iii*) in contrast to streaking, this method yields the energy dependence of the photoemission delay in a single measurement.

2. Results

In order to be able to study photoemission from the noble metal surfaces Ag(111) and Au(111), we introduced a refocusing toroidal mirror in the existing attosecond beamline [8]. This peculiar refocusing geometry offers the possibility to perform simultaneous measurements with two different, spatially separated targets. In this way, a well-known reference sample can be chosen to calibrate the experiment in one interaction region. For this experiment, a surface-science endstation equipped with a hemispherical electron analyzer was installed in the second interaction region. This combination enables measuring photoemission delays from a single initial state. We used few-cycle IR pulses with time duration between 7 and 12 fs to generate a comb of odd harmonics in a cell filled with argon. The two targets are ionized by the harmonics producing comb-like photoelectron spectra, which peak at the harmonic positions.

An example of simultaneously acquired RABBITT traces is shown in Fig. 1. The delayed IR pulse modulates the photoelectron spectrum. When the attosecond pulse train and the IR pulse overlap in time, satellites of the harmonic peaks (sidebands) appear in the recorded trace. The amplitude of these sidebands oscillates with twice the frequency of the IR beam. It can be shown that the phase of the photoelectron is recorded in the phase of the oscillatory signal. Thus, after subtraction of the calibration signal, one can access the surface specific photoemission phases displayed in Fig. 2.

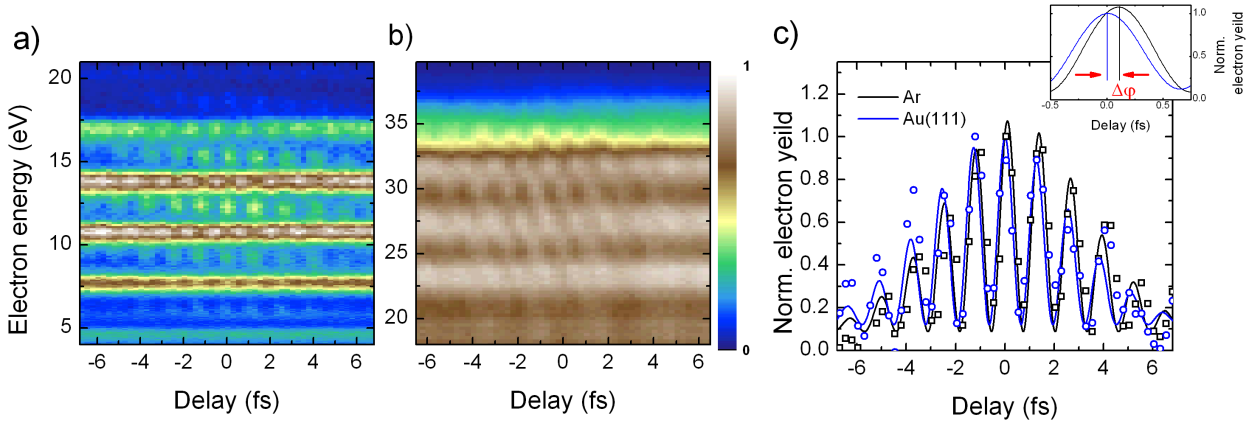

Fig. 1. Simultaneously recorded RABBITT traces for Ar *3p* and Au(111) *4d*-band. **(a)** and **(b)** show the experimental photoelectron spectra as a function of delay between the attosecond pulse train and the IR pulse. The two RABBITT traces are offset with respect to each other in energy due to the different ionization potential/work function of the respective targets **(c)** Shows the signal of sideband 18 extracted from (a) around 12.25 eV and (b) around 25.5 eV, black squares and blue rounds, respectively. The solid lines represent the fitting curve used to extract the experimental phases as is enlightened in the inset.

The energy dependence of the experimental phases is not monotonic. In particular the photoemission phases show a tendency to increase with energy. This is in contrast to the explanation of the previous results from the streaking experiments [6,7] where transport was assumed to be the dominant mechanism.

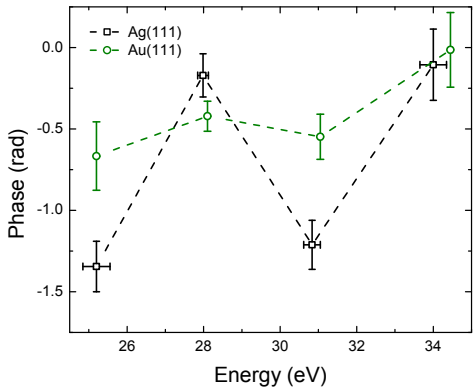

Fig. 2. Experimental surface specific phases for four different sidebands (SB 16, 18, 20, 22) for Ag(111) *4d*-band and Au(111) *5d*-band, black squares and green rounds, respectively.

3. Conclusion and outlook

In conclusion, we presented a new experimental approach that extends the RABBITT technique to condensed matter and accesses the surface specific response with the simultaneous acquisition of two traces. The experimental results revealed that a simple model dominated by transport is not suitable to describe the photoemission process in noble metals like Au(111) and Ag(111). It is worth to point out that the new detection scheme disposes of the requirement for a reference state in the second target and makes timing information for a single starting state accessible. Furthermore, a different scheme for the harmonic generation could allow a better energy resolution of the measurement and yield access to photoemission dynamics at other photon energies.

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