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# Atomic scale electronic structure and response in attosecond photoemission delays: A case study using non-centrosymmetric BiTeCl

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**Abstract.** Attosecond time-resolved photoemission from the differently terminated BiTeCl surfaces yield a photoelectron streaking that cannot be explained by bulk propagation effects alone. Instead, the atomic scale electronic structure and dynamical screening for both surface terminations have to be taken into account.

## 1 Introduction

Generation of intense attosecond light pulses and their application in photoelectron streaking spectroscopy allows capturing the photoemission process from different initial states with unprecedented temporal resolution [1]. Discrepancies between experimental observations and existing theoretical models advance our understanding of mechanisms that determine photoemission kinematics and, for example, allowed recently the identification of an intra-atomic delay as a significant contribution to the total photoemission delay [2]. Gathering further experimental evidence about attosecond time-resolved photoemission in different materials is essential to test and further expand our understanding as well as to pose new challenges for refined theoretical models.

In this case study we investigate attosecond time-resolved photoemission from the layered and non-centrosymmetric crystal BiTeCl. The lack of inversion symmetry allows

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studying photoelectron dynamics for differently terminated and well-defined inert surfaces. Furthermore, the different layer stacking enables the experimental determination of an important parameter, i.e. the inelastic mean free path (MFP) that critically affects the measured streaking delays. This reduces the ambiguities of streaking spectra analysis and thus allows identifying additional atomic scale effects that influence the streaking.

## 2 Experimental Setup

The photoelectrons are excited by an isolated extreme ultraviolet (EUV) pulse (center photon energy $E_{EUV} \approx 91\,\text{eV}$) and probed by a few-cycle IR-pulse ($E_{IR} \approx 1.55\,\text{eV}$). A field-free time-of-flight spectrometer is used as photoelectron detector with a small full acceptance angle ($\approx 6°$) which guarantees the predominant detection of photoelectrons emitted in normal direction. The magnetic fields were compensated by Helmholtz coils. To ensure longer measuring times on clean surfaces all crystals were cleaved and studied under UHV conditions ($\approx 7 \cdot 10^{-11}$ mbar).

## 3 Results and Discussion

The EUV photoemission spectrum is composed of emission peaks related to the valence band, as well as the Bi-5d and Te-4d core levels. In fig. 1 the extracted relative delays $\Delta t_{Bi\text{-}Te}$ between the core level states Bi-5d and Te-4d are summarized for the differently terminated surfaces and samples.

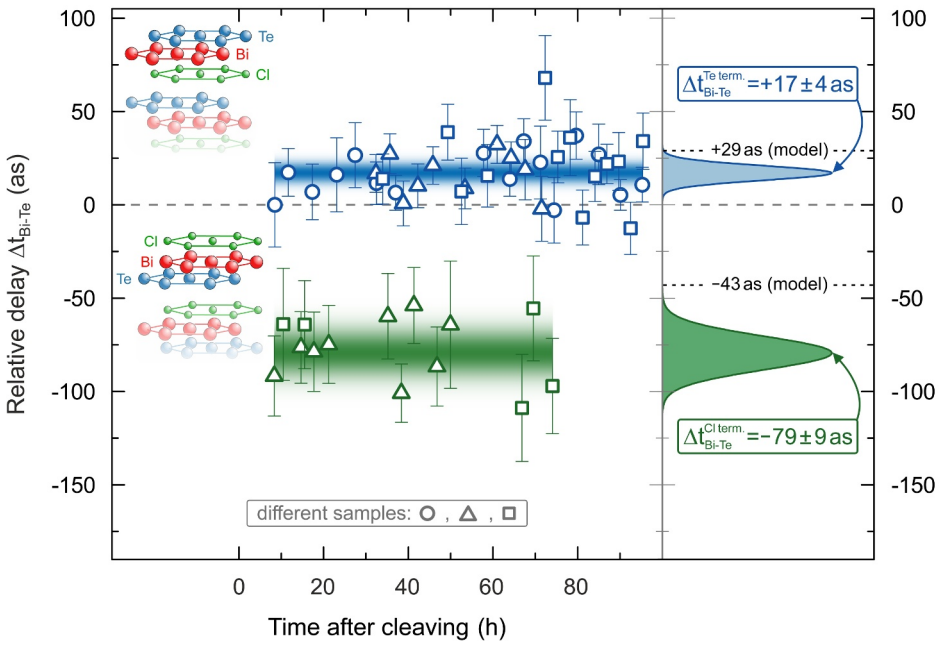


**Fig. 1.** Left panel: Experimentally observed relative streaking delays between the emissions from the core levels Bi-5d and Te-4d as a function of the time after cleaving for both surface terminations (Te-term.: blue, Cl-term.: green) and different samples (○, △, □). Right panel: Comparison between the averaged delays from the left panel and the results from electron trajectory calculations (dashed lines).

After the cleaving process, the streaking measurements were recorded over 80 hours without any sign of systematic drift. By repeated measurements on different samples and sample positions the experimental uncertainty of $\Delta t_{Bi\text{-}Te}$ was reduced to ±4 as for the Te-terminated surface. In case of the Te-surface the Bi-5d electrons are delayed by 17±4 as with respect to the Te-4d electrons. This situation changes dramatically for the Cl-terminated surface. In this case $\Delta t_{Bi\text{-}Te}$ changes sign because of the inverse layer stacking, and now the Te-4d electrons are delayed by 79±9 as with respect to the Bi-5d electrons.

The comparison with results from classical and quantum mechanical electron trajectory calculations results in a significant discrepancy for both terminated surfaces. The modeling includes the experimentally observed MFP (≈3.5 Å), the screened electron-hole interaction using a Yukawa potential with a screening length of $\mu$=5 Å, an inner potential with the step height $U_{IP}$=9.5 eV [3,4], and a penetrating IR-field inside the material (transmission coefficient $\tau_0 \approx 0.126$, extinction coefficient $\kappa \approx 8.15 \cdot 10^{-4}$ Å$^{-1}$, optical data from [5]). Additionally, the influence of intra-atomic corrections within the Eisenbud-Wigner-Smith theory was taken into account, but in case of the Bi-5d and Te-4d electrons this additional delay is negligible small. For the Te-terminated surface the model delivers a relative delay of +29 as and for the Cl-terminated surface a delay of −43 as.

The discrepancy between model and experiment is rather striking. Preliminary model calculations considering atomic scale variations of the streaking field distribution indicate that this degree of freedom significantly affects absolute and relative streaking delays. For example, the observed variation of the streaking amplitude for different surface terminations and emission channels can only be explained if atomic scale variations in the IR streaking field are considered. Bulk propagation properties such as mean free path, effective mass or continuum band structure effects of the material are independent of surface termination and thus have to be chosen identical for both terminations. Even with a broad variation of these parameters alone the observed streaking cannot be explained. Hence we conclude that the discrepancy between model and experiment arises from atomic scale differences at the two surfaces either with respect to the electronic configuration and/or the dynamic screening of the IR streaking field. This demonstrates that a proper theoretical model must account for the details of the electronic structure and atomic scale dynamical screening at the interface.

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