

Comparative time-resolved photoelectron spectroscopy from Cu(100) and Cu(111) surfaces

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Synopsis We study the effects of initial-state modeling and Fresnel reflection of the incident IR pulse on time-resolved IR-laser-assisted XUV photoelectron spectroscopy of Cu(100) and Cu(111) surfaces. These effects are shown to strongly influence the photoelectron spectra, and thus need to be incorporated in theoretical calculations. Comparative RABBITT phases are discussed as a way of eliminating the phase shifts introduced by the unknown high harmonic phases of the XUV pulse train.

We numerically obtain RABBITT (reconstruction of attosecond beating by interference of two-photon transitions) [1] spectra for photoemission from Cu(100) and Cu(111) surfaces (Fig. 1). From the spectra we extract sideband (SB) phase shifts that we analyze with regard to the (i) localization character of the initial photoelectron (PE) states, (ii) initial state momentum distributions, (iii) depth-dependent contributions to the dipole transition matrix element and photoemission yield, and (iv) electric-field amplitude distribution of the assisting IR field near the surface and inside the substrate [2].

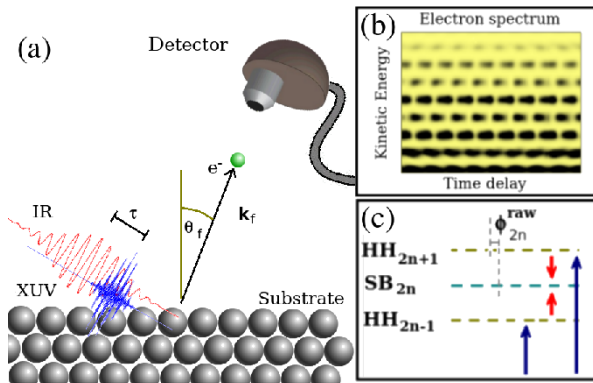


Figure 1. Schematic of the RABBITT method: Single-photon absorption in an XUV pulse train followed or preceded by photon absorption or stimulated emission in a delayed IR pulse.

We represent the electronic initial valence-band based on effective single-electron model potentials [3] and we determine the band limits of occupied initial states from recently obtained XUV photoelectron spectra in Ref. [4]. For the final photoelectron state we use a modified Volkov state [2], allowing for (i) the space dependence of the IR electric field induced by the reflection/transmission at the surface and (ii) an overall exponential decay inside the solid that accounts for electronic collisions in the substrate.

For the analysis of the spectra we remove the *static* PE background given by the XUV-only ioniza-

tion from the XUV-plus-IR spectra (Fig. 2).

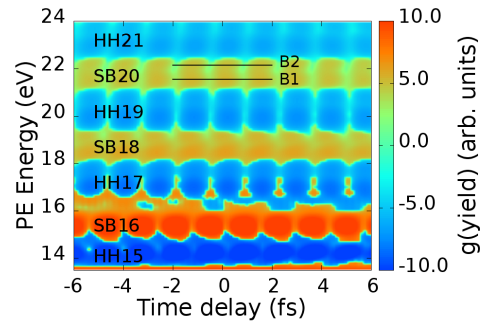


Figure 2. XUV-subtracted RABBITT spectrum from Cu(111) showing a distorted image of the initial-state density.

We find (a) the localization character of the initial states to significantly influence RABBITT spectra and (b) that it is necessary to include the surface-reflection of the incident IR-pulse. We model the IR-pulse reflection based on the dielectric properties of the substrate [5] and scrutinized its effects for emission from (i) the surface state of Cu(111) and (ii) bulk valence states of Cu(111) and Cu(100). In addition, we discuss Cu(111)-Cu(100) RABBITT phase differences as a means to eliminate the unknown HH phases.

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References

- [1] R. Locher et al. 2015 *Optica* **2**, 405
- [2] M. J. Ambrosio et al. 2016 *Phys. Rev. A* **94**, 063424
- [3] E. V. Chulkov et al. 1999 *Surf. Sci.* **437**, 330
- [4] F. Roth et al. 2016 *J. Electron Spectrosc.* **208**, 2
- [5] A. D. Rakić et al. 1998 *Appl. Opt.* **37**, 5271

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