Time-resolved photoemission interferograms of Cu, Ag, and Au surfaces

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Attosecond pulse trains that are synchronized with the driving IR laser allow for time-resolved investigations of the electronic dynamics in matter by reconstruction of attosecond beating by interference of two-photon transitions (RABBITT) [1-5].

RABBITT spectra and phases from Ag(111) and Au(111). Figure 1 shows our numerical results obtained by representing the d-valence band of the target by tight-binding states [1]. We found spectra calculated with tight-binding initial states to agree better with the experimental spectra in [4,5] than a DFT-based (“Chulkov” potential) modeling of the valence band. Fresnel reflection of the incident IR pulse enhances side-band yields and induces a harmonic-order-independent phase shift of -1.6 radians.

**Fig. 1:** Experimental [4] (a) and simulated [1] (b) RABBITT spectra for Au(111). (c) RABBITT phases relative to Ar gas phases. Experimental phases [4] include a beam propagation phase.

We predict distinctly different RABBITT spectra from Cu(111) and Cu(100) [2] (Fig. 2). These can be distinguished experimentally with existing technology (Fig. 2a). As Fresnel reflection reduces the IR skin depth, we find the inclusion of the Fresnel-reflected incident IR pulse (i) prerequisite for reproducing the measured Cu(111) RABBITT in [5] and (ii) to modify photoelectron spectra from bulk and surface states differently, revealing their different degrees of spatial localization [2].

**Fig. 2:** (a) Suggested in situ comparative RABBITT setup [1] placing the two surfaces on a sliding platform while keeping all path lengths fixed. Spectra after subtraction of the single-XUV-photon yield for (b) Cu (100) and (c) Cu(111). (d) Sub-bands “B1” and “B2” of the Cu (111) valence band [1] are imaged in (c).


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