RABBITT spectroscopy of transition-metal surface states

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Synopsis We distinguish contributions to the RABBITT phases from surface states of transition metals, due to Fresnel-reflection of the incident IR pulse, the high harmonics of the XUV pulse-train and the XUV photo release process.

We present numerical RABBITT (reconstruction of attosecond beating by interference of two-photon transitions) [1] spectra for photoemission from solids, the occupied surface states of Cu(111), Au(111), and Ag(111) (Fig. 1). We generate the transition-metal surface states based on an effective single-electron potential capable of reproducing the main characteristics of the substrate valence-band electronic surface [2].

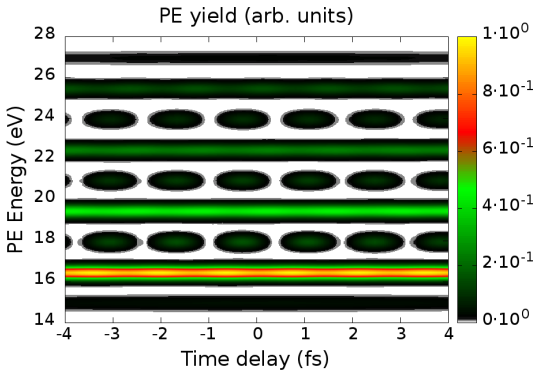

Figure 1. RABBITT spectrum from the Au(111) surface state.

Our numerical spectra show RABBITT phases to be strongly affected by the IR pulse Fresnel reflection at the solid-vacuum interface [3], which introduces a phase due to the photoelectron propagating through the overall (i.e., incident plus reflected) electric field. This Fresnel phase ϕ_E largely determines the raw RABBITT phases

$$\phi_{2n}^{RAB} \equiv \phi_{2n-1}^{+} - \phi_{2n+1}^{-} - \phi_{2n-1}^{(HH)} + \phi_{2n+1}^{(HH)} + 2\phi_E, \quad (1)$$

as shown in Fig. 2 for different incident directions and detection angles. Since it is well determined, the Fresnel phase contribution $2\phi_E$ can be subtracted from the raw RABBITT phase ϕ_{2n}^{RAB}, therefore leaving only the contributions from the HH phases $\phi_{2n-1}^{(HH)}$ and $\phi_{2n+1}^{(HH)}$, of the XUV pulse train, and the scattering phases $\phi_{2n-1}^{+} - \phi_{2n+1}^{-}$ that are related to the ionization process. ϕ_{2n-1}^{+} corresponds to the absorption of an XUV photon of energy $(2n-1)\omega_{IR}$ plus the absorption of an IR photon, while ϕ_{2n+1}^{-} stems from the absorption of an XUV photon with energy $(2n+1)\omega_{IR}$ plus the release of an IR one. These scattering phase differences in our calculations are small, due to the very short propagation time of the photoelectron in the substrate.

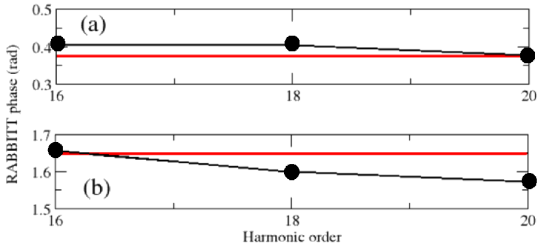

Figure 2. RABBITT phases from the Ag(111) surface state (dots) and $2\phi_E$ (red line). (a) Incidence angle 30°, with the XUV and IR polarizations tilted 30° from the incidence plane. Electron emission at 28° from the surface normal and 18° from the emission plane. (b) Incidence angle 80°, with the polarization axis tilted 20° from the incident plane, for normal emission.

Thus,

$$\phi_{2n}^{RAB} - 2\phi_E \approx -\phi_{2n-1}^{(HH)} + \phi_{2n+1}^{(HH)} \quad (2)$$

for the odd harmonics in the XUV pulse train can be used to estimate the unknown high harmonic phases $\phi_{2n+1}^{(HH)}$.

Acknowledgements

We acknowledge support from NSF grant PHY-146 4417, the A. v. Humboldt foundation, and the Division of Chemical Sciences, Office of the Basic Energy Sciences, Office of Energy Research, US DoE.

References

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