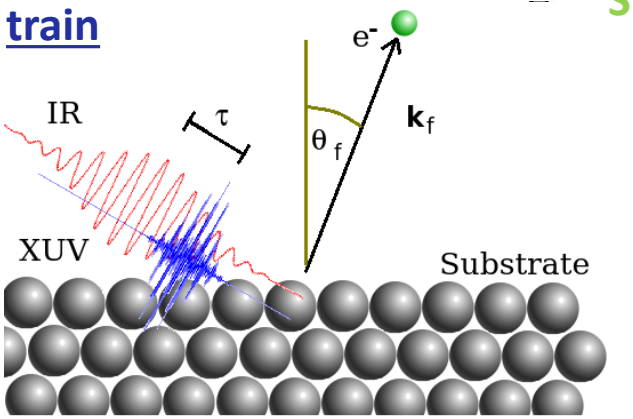
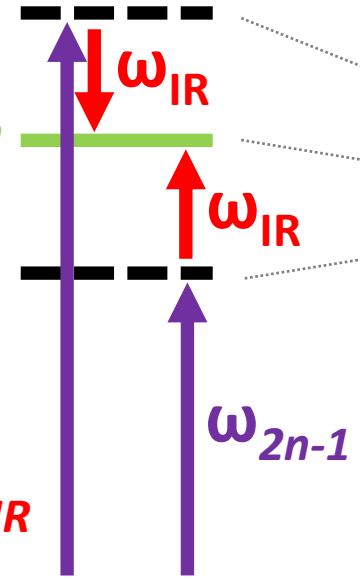
# RABITT spectra from surfaces

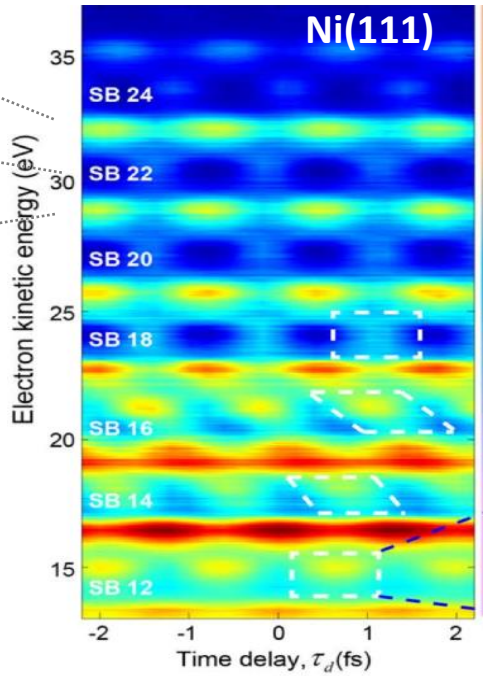
(Reconstruction of Attosecond Beating By Interference of Two-photon Transitions)

**Experiment:** Tao *et al.*, *Science* **353**,62 (2016)

incident **attosecond XUV-pulse train**




Photoelectron yield: $$P(\mathbf{k}_f, \tau) \propto P_0 + P_1 \cos\left(2\omega_{IR}\tau - \phi_{2n}^{RAB}\right)$$

RABBITT phase shifts: $$\phi_{2n}^{RAB} \equiv \phi_{2n-1}^{+} - \phi_{2n+1}^{-} - \left(\phi_{2n-1}^{(HH)} + \phi_{2n+1}^{(HH)}\right)$$

unknown HH phases

Chen, U.T., Murnane, *et al., PNAS* **114**, E5300 (2017)

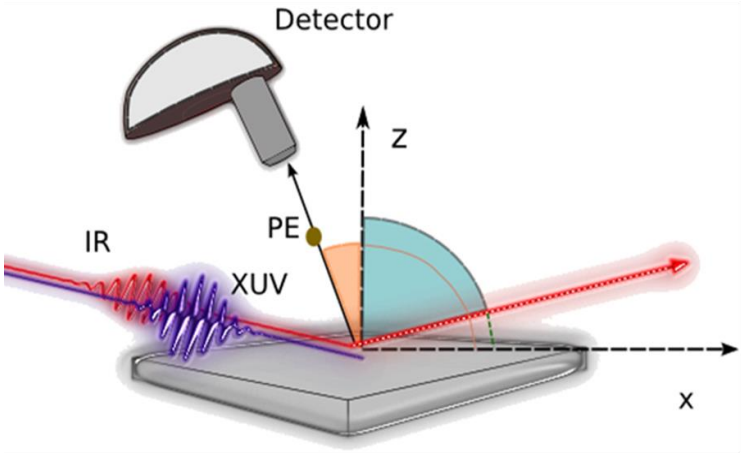
# RABITT spectra from surfaces

Attosecond pulse trains synchronized with and delayed (or advanced) relative to 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) [1-6]. We found spectra calculated with tight-binding initial states to agree better with the experimental spectra in [1,2] than a DFT-based ("Chulkov" potential) [7] 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. We predict distinctly different RABBITT spectra from Cu(111) and Cu(100) [5], which can be distinguished experimentally with existing technology. As Fresnel reflection reduces the IR skin depth, we find the inclusion of the Fresnel-reflected incident IR pulse (i) prerequisite for reproducing measured Cu(111) RABBITT spectra [2] and (ii) to modify photoelectron spectra from bulk and surface states differently, revealing their different degrees of spatial localization [5].

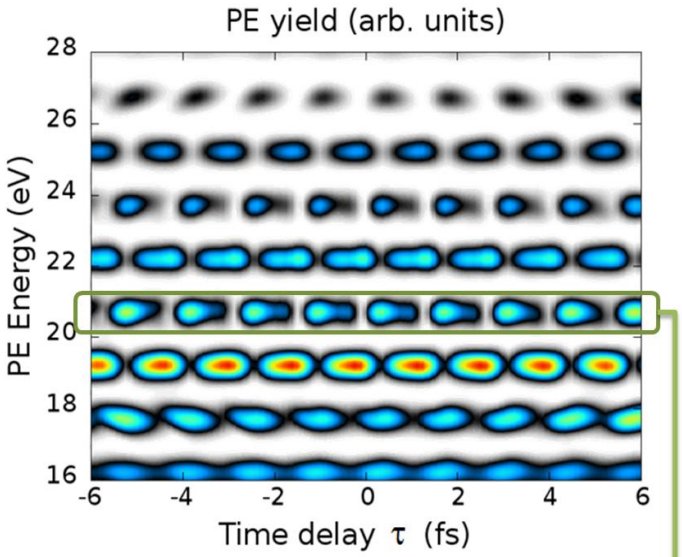

[1] R. Locher *et al*. 2015 Optica **2**, 21323.
[2] M. Lucchini et al. 2015 Opt. Express **23**, 8867.
[3] Z. Tao *et al*. 2016 Science **353**, 62.
[4] C. Chen *et al.* 2017 Proc. Natl. Acad. Sci. **114**, E5300.
[5] M. J. Ambrosio *et al.* 2016 Phys. Rev. A **94**, 063424.
[6] M. J. Ambrosio *et al.* 2017 Phys. Rev. A **96**, 051403(R).
[7] E. V. Chulkov *et al*. 1999 Surf. Sci. **437**, 330.

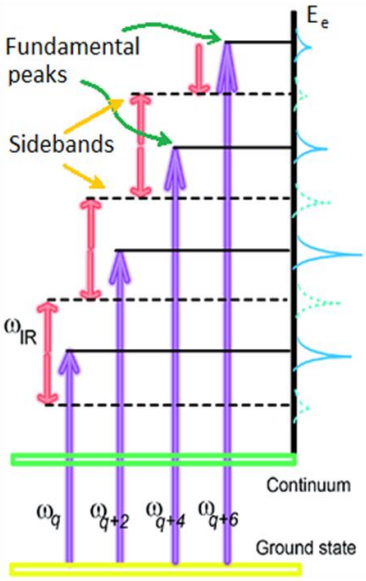
# RABITT spectra from surfaces

## Basic setup



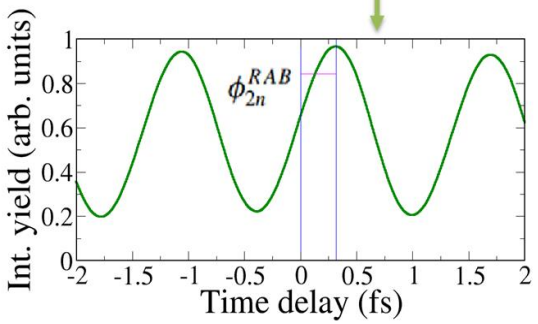

## Spectrum and energetics




## Raw RABBITT phases:

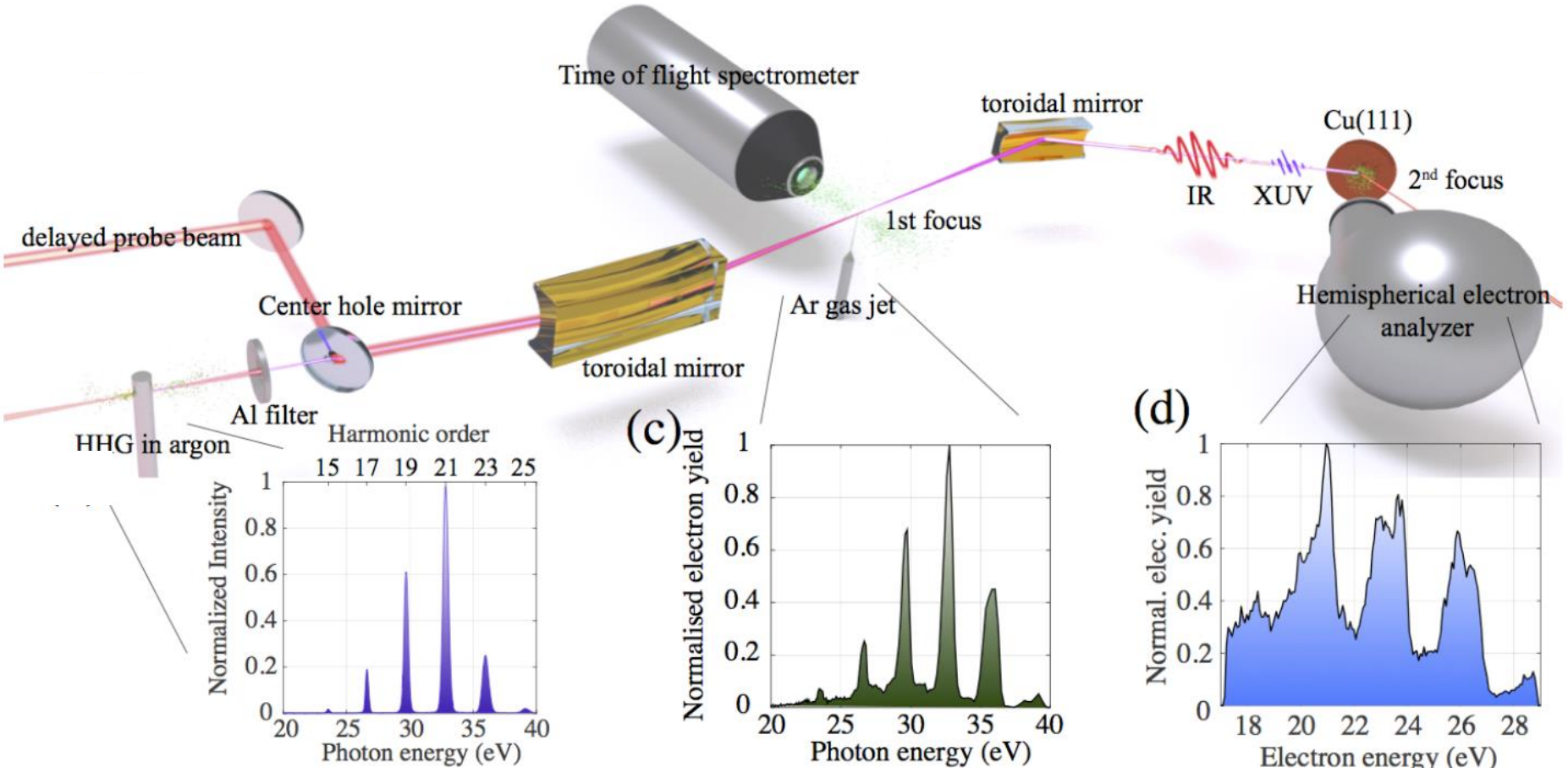
$$\phi_{2n}^{RAB} \equiv \underbrace{\phi_{2n-1}^{+} - \phi_{2n+1}^{-}}_{\text{Scattering phases}} - \underbrace{\phi_{2n-1}^{(HH)} + \phi_{2n+1}^{(HH)}}_{\text{HH phases}}$$



# RABBITT spectra from surfaces

RABBITT phase shifts: $\phi_{2n}^{RAB} \equiv \phi_{2n-1}^{+} - \phi_{2n+1}^{-} - \phi_{2n-1}^{(HH)} + \phi_{2n+1}^{(HH)}$

unknown HH phases



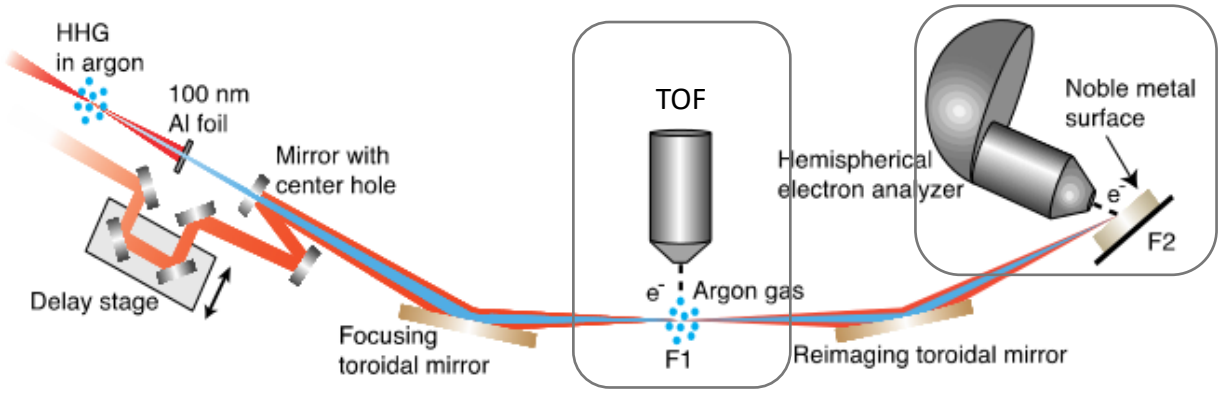

Kasmi, Keller, *et al.,* Optica **4**, 2334 (2017)

# RABBITT spectra from surfaces

RABBITT phase shifts: $\phi_{2n}^{RAB} \equiv \phi_{2n-1}^{+} - \phi_{2n+1}^{-} - \left(\phi_{2n-1}^{(HH)} + \phi_{2n+1}^{(HH)}\right)$ unknown HH phases

**Remove the HH phases**

Comparison with atomic target



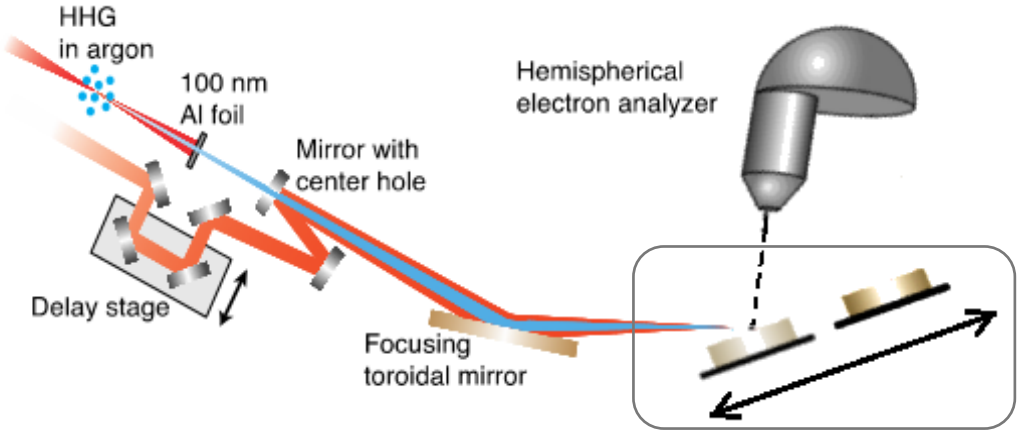

Locher, Keller, *et al*. Optica 2, 21323 (2015)

Proposed alternative:

**Sliding platform**

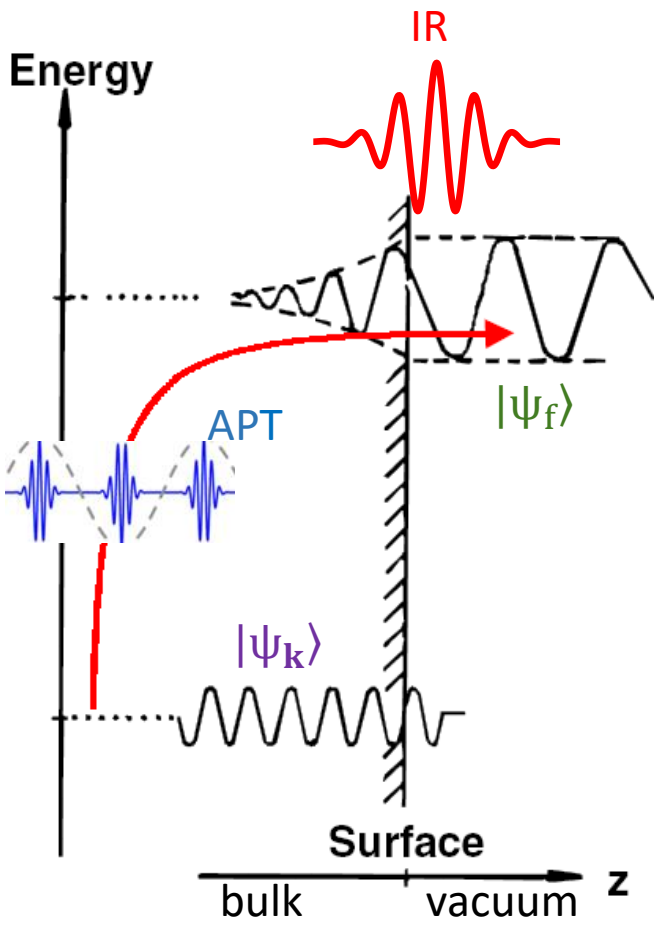
Advantage:

Invariant optical path lengths




Ambrosio, U.T., *PRA* **94**, 063424 (2016)

# Time-resolved interferometric photoemission from metal surfaces




$$T_k \sim \int_{-\infty}^{\infty} dt \, \langle \psi_f(t) | \, \mathbf{p} \cdot \mathbf{A}_{XUV}(t+\tau) \, | \psi_k(t) \rangle \qquad \text{amplitude}$$

$$P = \sum_{\mathbf{k} \in 1.BZ} |T_k|^2 \qquad \text{probability}$$

**Initial state**: Bloch wave

$$\psi_k(\mathbf{r}, t) \sim e^{-i E_k t + i \mathbf{k} \cdot \mathbf{r}} \; u_k(\mathbf{r}) \quad + \text{refl. wave}$$

$$u_k(\mathbf{r}) = u_k(\mathbf{r} + \mathbf{R}_n)$$

**Final state**: damped Volkov wave

$$\psi_f(\mathbf{r}, t) \sim e^{i [\mathbf{k}_f + \mathbf{A}_{IR}] \cdot \mathbf{r} \; + \; i \, \phi_V(\mathbf{k}_f, t)}$$

$$k_{f,z} = Re\{k_{f,z}\} - i / [2 \lambda(k_f)] \qquad \text{damping factor}$$

$$\phi_V(\mathbf{k}_f, t) = \frac{1}{2} \int_t^{\infty} d\tau \; |\mathbf{k}_f + \mathbf{A}_{IR}(\tau)|^2 \qquad \text{Volkov phase}$$

Ambrosio, U.T., PRA **94**, 063424 (2016)

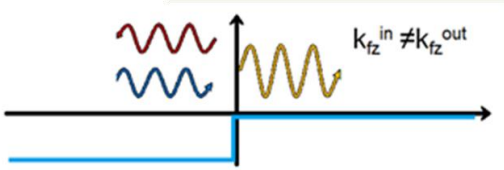
# Basic theory

**Transition matrix:**

**Final state**: damped Volkov function

$$\Psi_{\mathbf{k}_f}^{f}(\mathbf{r},t,\tau)=\frac{f_{\varepsilon_f,\theta_f}(z)}{(2\pi)^{3/2}}e^{i\mathbf{k}_{f,\parallel}\cdot\mathbf{r}_{\parallel}}\psi_{k_{f,z}}(z)e^{-i\phi_{\mathbf{k}_f}(\mathbf{r},t)}e^{-i\varepsilon_{\mathbf{k}_f}t}$$

$$f_{\varepsilon_f,\theta_f}(z)=\Theta(z)+e^{z/[2\lambda(\varepsilon_f)\cos(\theta_f)]}\Theta(-z)$$




$$\mathbf{A}_{IR}(\mathbf{r},t)=\int_t^{\infty}\mathbf{E}_{IR}(\mathbf{r},t')\,dt'$$

$$\phi_{\mathbf{k}_f}(\mathbf{r},t)=\int_t^{\infty}dt'\mathbf{k}_f\cdot\mathbf{A}_{\mathbf{IR}}(\mathbf{r},t')$$

**Initial state** (translationally invariant):

$$\Psi_{\mathbf{k}_i}(\mathbf{r},t)=\frac{e^{-i\varepsilon_{\mathbf{k}_i}t}}{(2\pi)}e^{i\mathbf{k}_{\parallel}\cdot\mathbf{r}_{\parallel}}\psi_i(z)$$

**Net IR-electric-field:** superposition of incident and reflected IR field, calculated from Fresnel's equations for a complex Lorentz-Drude dielectric function [8].

$$\mathbf{E}_{IR}(\mathbf{r},t)=\mathcal{E}_{IR}(t)\,[\;\mathbf{E}_{ext}(z)\mu(z)+\mathbf{E}_{in}(z)\mu(-z)]$$

$$\mu(z)=\frac{1}{2}\left[\tanh\left(\frac{z}{a_s}\right)+1\right]$$

$$[\mathbf{E}_{ext}(z)\mu(z)+\mathbf{E}_{int}(z)\mu(-z)]=|\mathbf{E}_{ext}(z)\mu(z)+\mathbf{E}_{int}(z)\mu(-z)|\,e^{i\phi_E(z)}$$

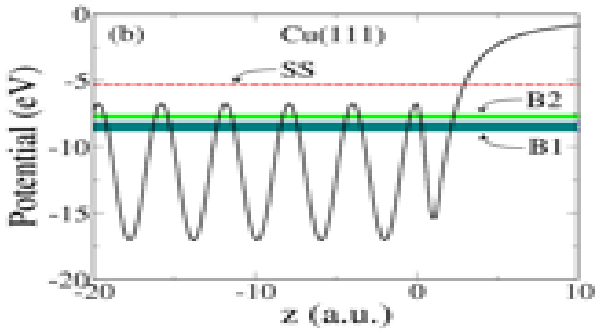
**XUV pulse train:**

$$\mathbf{E}_{XUV}(\mathbf{r},t)=\sum_n\mathbf{E}_{0,2n+1}e^{-\frac{(\omega-\omega_{2n+1})^2}{2\sigma_{2n+1}^2}\log(2)}\cos\left(\omega_{2n+1}t+\phi_{2n+1}^{HH}\right)$$

Unknown HH phases

# Initial states $\psi_i(z)$

## Chulkov potential



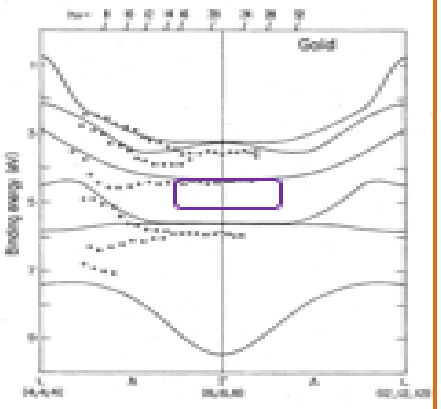

Effective potential (Chulkov), adjusted to DFT calculations [7]. B1/B2: occupied valence states according to Ref. [9] for (a) Cu(111).

## Tight binding model

- Highly localized d-band [10] motivates the use of a tight-binding representation.
- Flat dispersion relation: zero bandwidth model

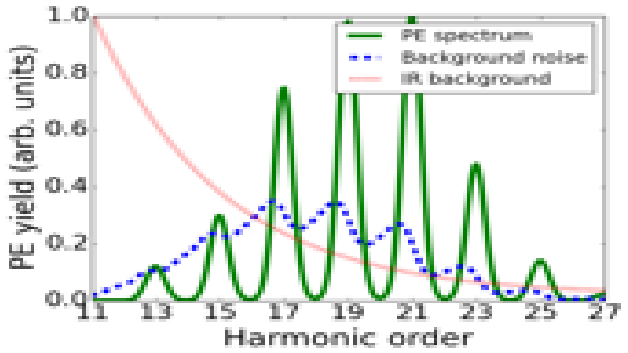
z-dependent orbitals:

$$\phi_i(z) = \sum_j e^{i\vec{k}_{\parallel,i}\cdot\vec{r}_j}\varphi_{n,\ell,m}(z - z_j)$$




Au(111) band structure [11]

## IR background and secondary electrons

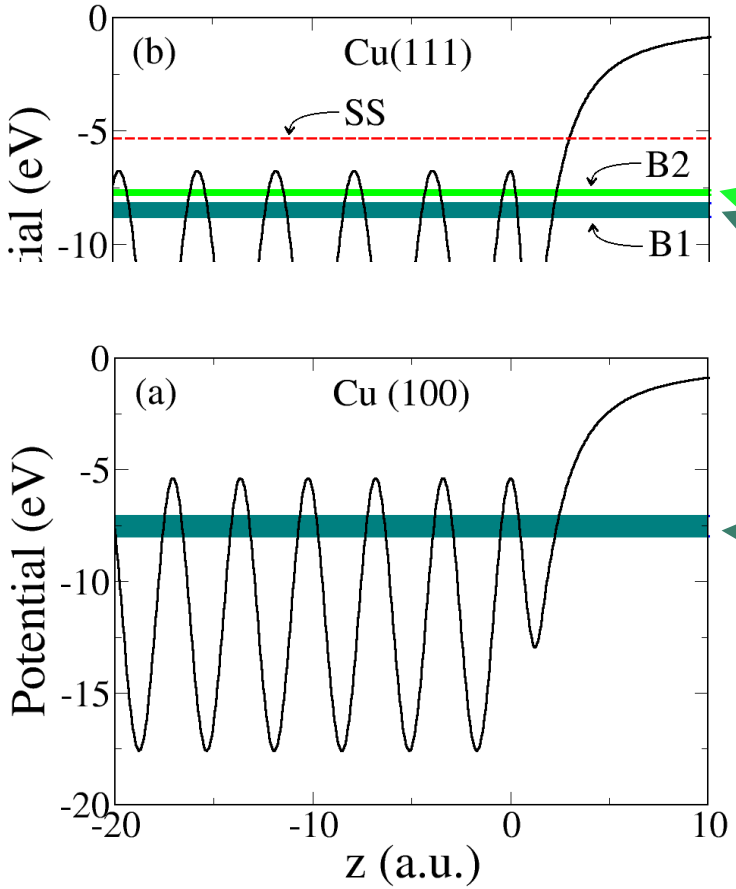


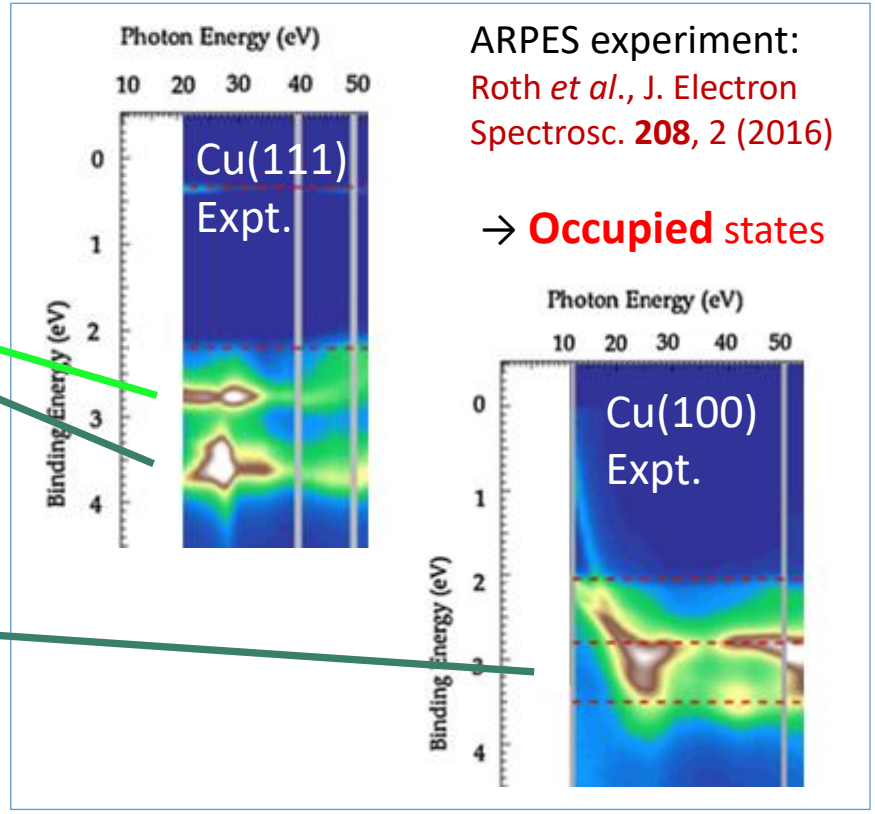

- Secondary electrons are promoted to the continuum by the XUV pulse train, but subsequently scattered inside the substrate. For this we devised a phenomenological model to account for losses due to electron-electron collisions.
- IR contribution from Locher *et al.* [1]. Fitting it with an exponential decay produced a good approximation within the energy range of interest.

# Surface electronic structure

"Chulkov potential" (LDA fit)

$$\Psi^{i}_{\mathbf{k}_i}(\mathbf{r},t) = \frac{e^{-i\varepsilon_{\mathbf{k}_i}t}}{2\pi} e^{i\mathbf{k}_{i\parallel}\cdot\mathbf{r}_{\parallel}} \psi_i(z)$$



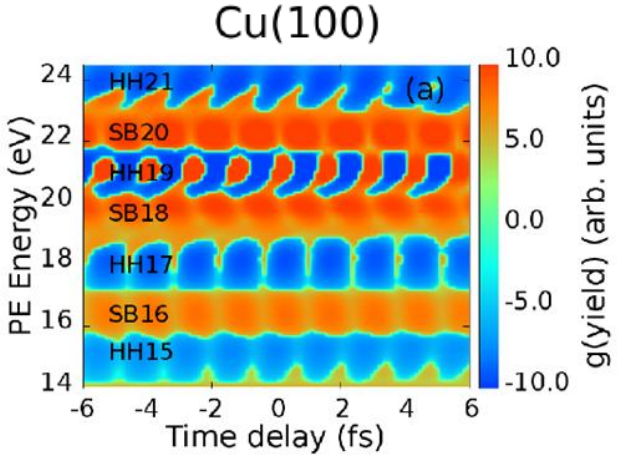

ARPES experiment:
Roth *et al.*, J. Electron Spectrosc. **208**, 2 (2016)

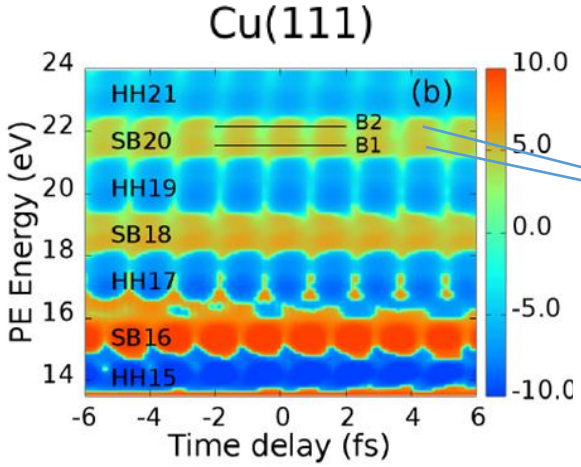
→ **Occupied** states

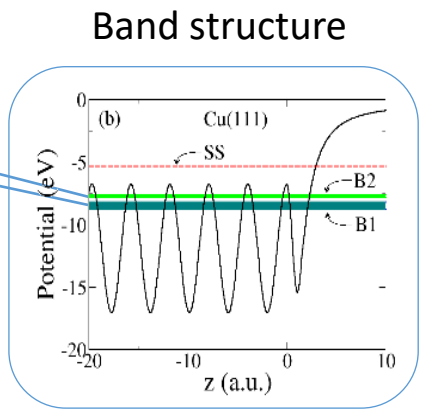
Ambrosio, U.T., PRA **94**, 063424 (2016)

# RABBITT spectra from Cu surfaces

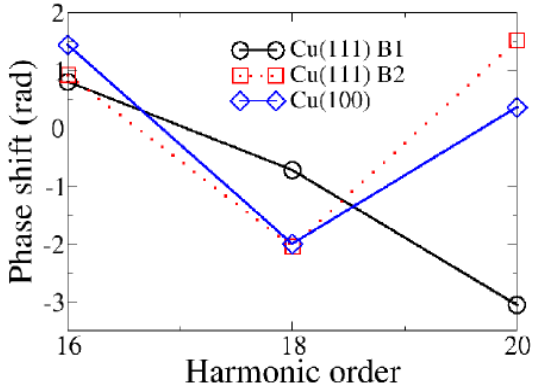
XUV-background subtracted



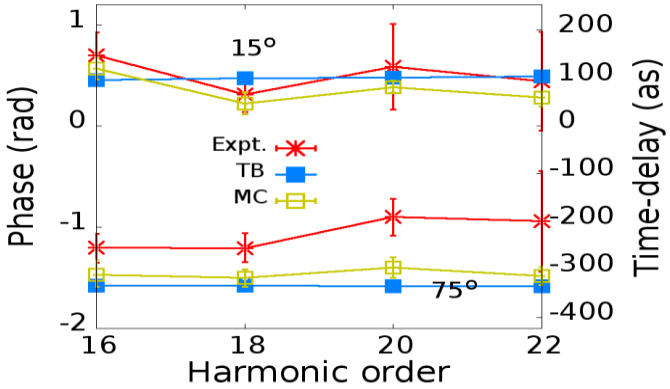




## Cu RABBITT phases relative to Cu(111)surface state




- Fresnel reflection matters
- IR pulse attenuation affects surface states less than bulk states

## Cu(111) RABBITT phases relative to gaseous Ne (15° and 75° incidence)



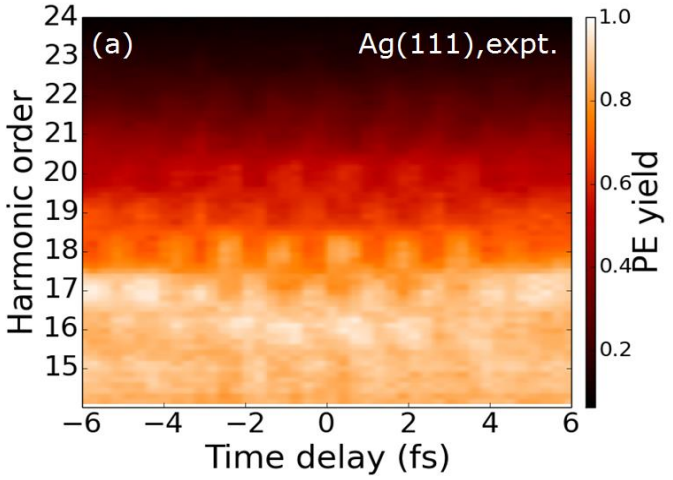

**TB:** tight binding model
**MC:** class. Monte Carlo model

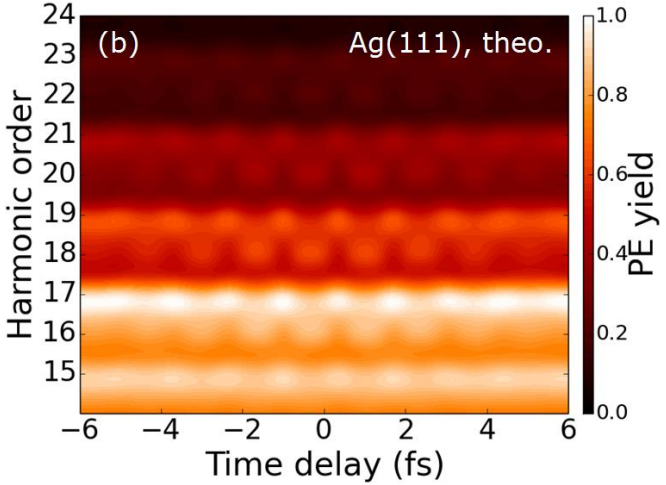
**Theory:** Ambrosio, U.T., *PRA* **94**, 063424 (2016); *PRA* **96**, 051403 (2017)
**Expt.:** Lucchini, Keller, *et al.*, *PRL* **115**, 137401 (2015)

# RABBITT spectra from Ag(111) surfaces

**Expt:**
Locher, Keller *et al.*, *Optica* **2**, 21323(2015)

**Theory:** *Tight binding. Gen. Sturmian basis.*
Ambrosio, U.T. in prep.




## RABBITT phases relative to Ar gas target

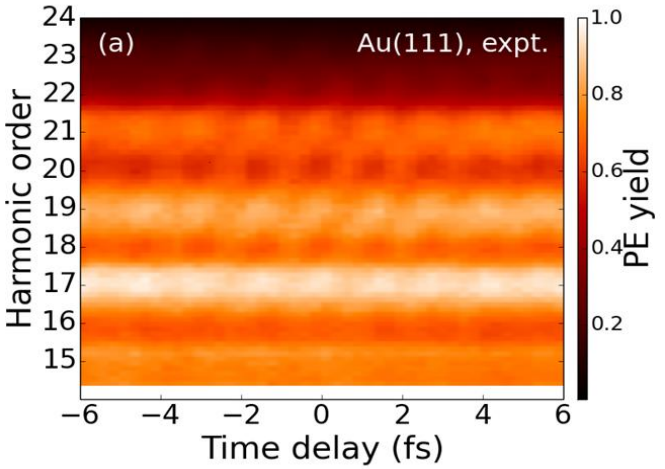
- Ar RABBITT phases calculated by Mauritsson *et al., PRA* **72**, 013401 (2005) subtracted from our calculated Ag(111) phases.
- Calculated RABBITT spectrum includes a delay independent IR ATI and XUV background.




Ambrosio, U.T., *PRA* **97**, 043431 (2018)

# RABBITT spectra from Au(111) surfaces

**Expt:**
Locher, Keller *et al.*, *Optica* **2**, 21323(2015)

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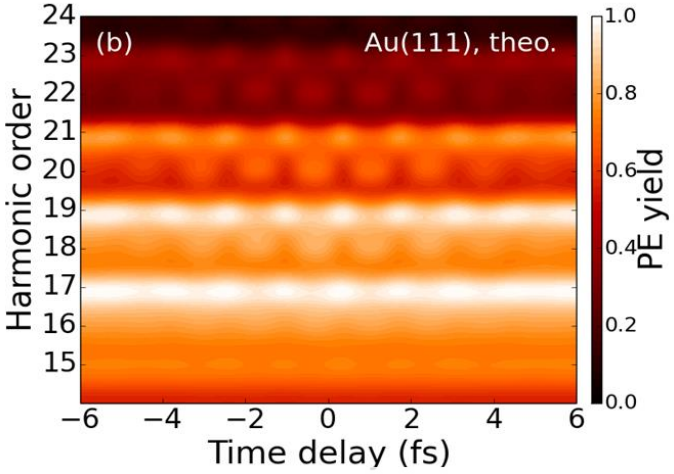


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Ambrosio, U.T., *PRA* **97**, 043431 (2018)