Description and influence of the IR probe field in the attosecond spectroscopy of solid surfaces

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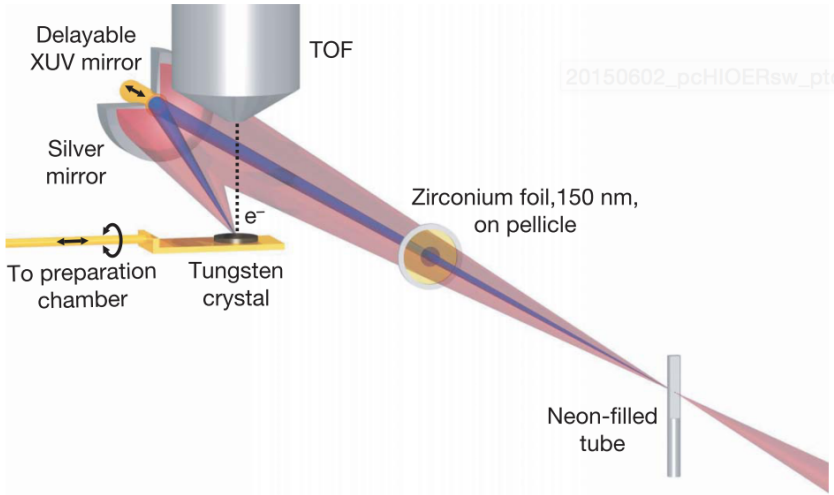
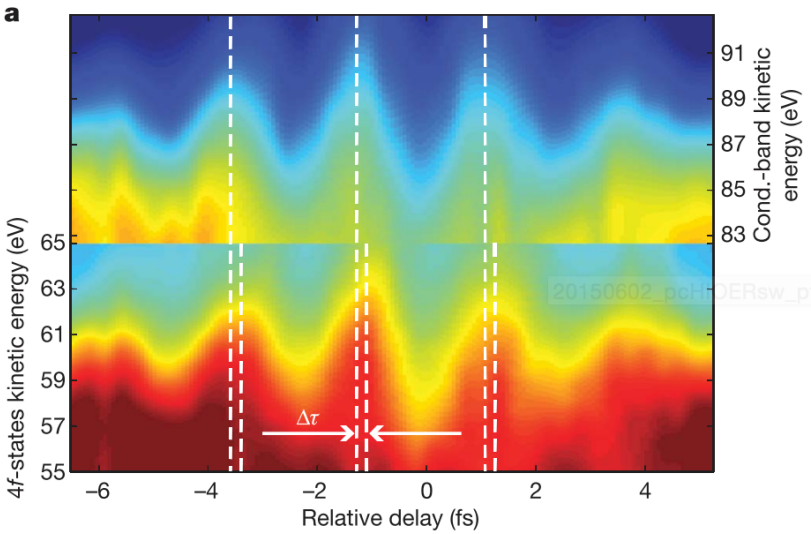
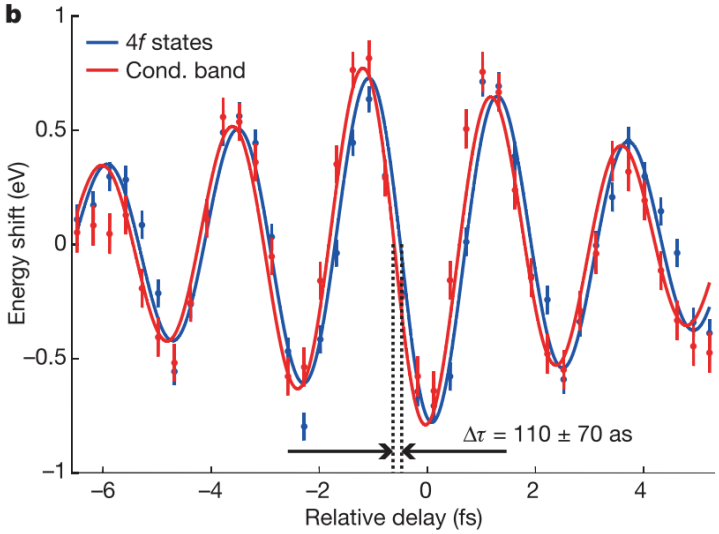
Relative delays in photoemission from W(110)







Attosecond streaking

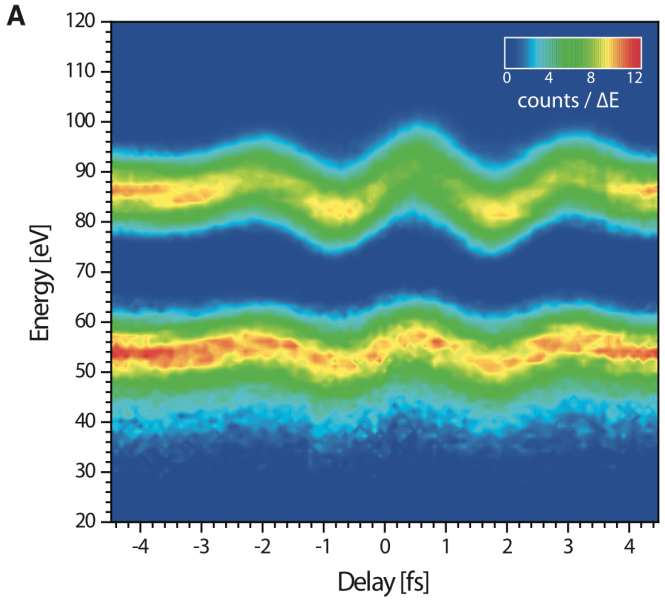
Relative delay between 4f and VB

$\Delta\tau = 110 \pm 70$ as

Cavalieri et al. Nature. 449, 1029 (2007)

Delays in photoemission from atoms

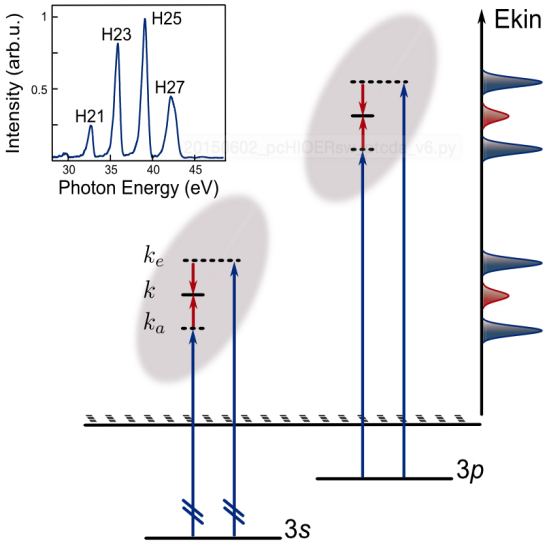
Streaking with SAP in Ne



Relative delay $\Delta\tau = 21 \pm 5$ as
between 2s and 2p in Ne

Single attosecond pulses (SAP)
with a delayed few-cycle IR field
lead to a time-dependent variation
of a photoelectron momentum

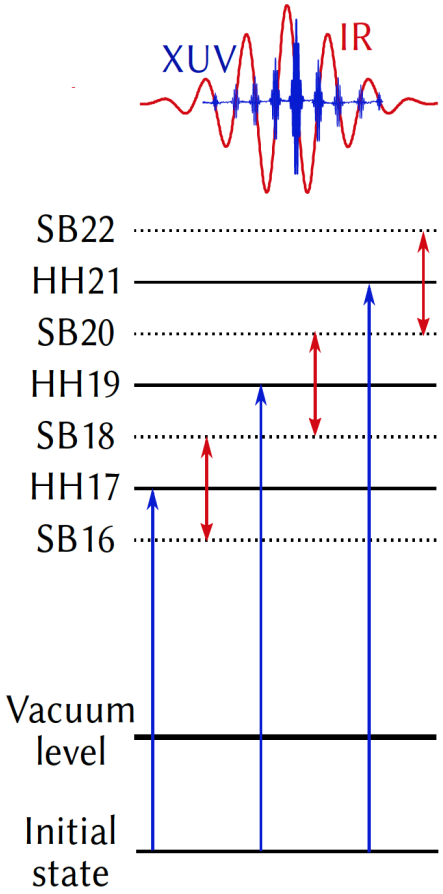
RABBITT with APT in Ar

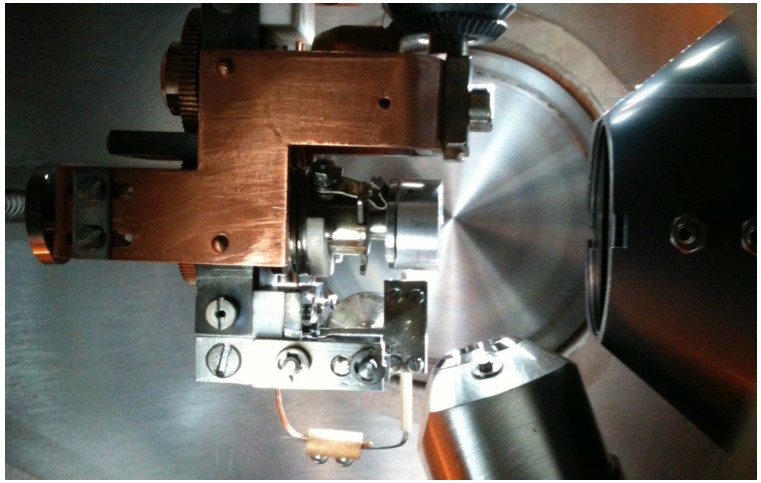


Relative delay $\Delta\tau = 30...120$ as
between 3s and 3p in Ar

Attosecond pulse trains (APT)
with a delayed few-cycle IR field
lead to a formation of sidebands
due to quantum path interference

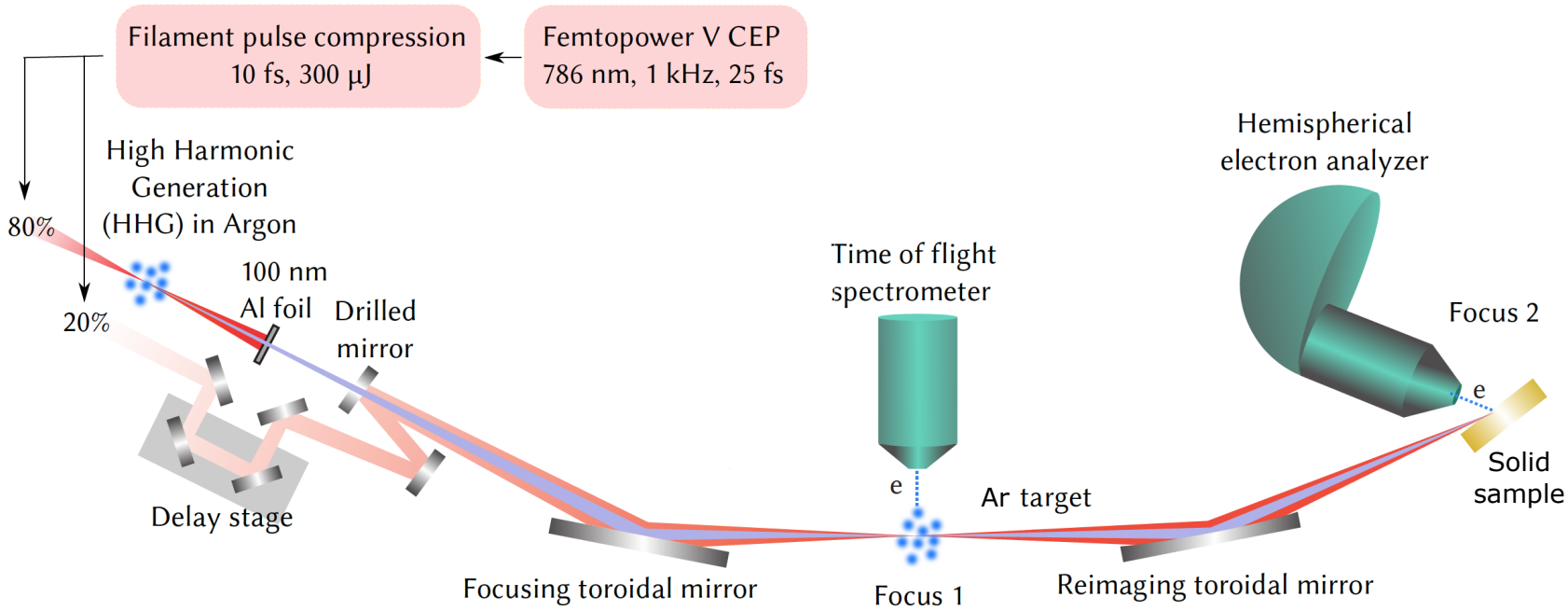
M. Schultze et al. Science 328, 1658 (2010), Kluender et al. PRL 106, 143002 (2011)

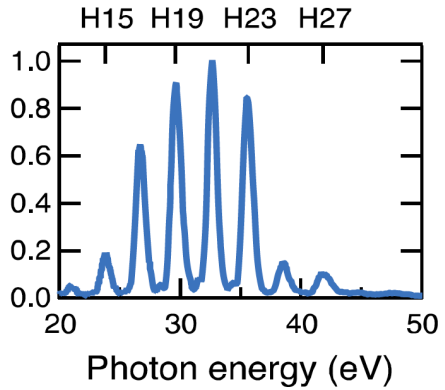




Extension of RABBITT technique
to solid-state targets

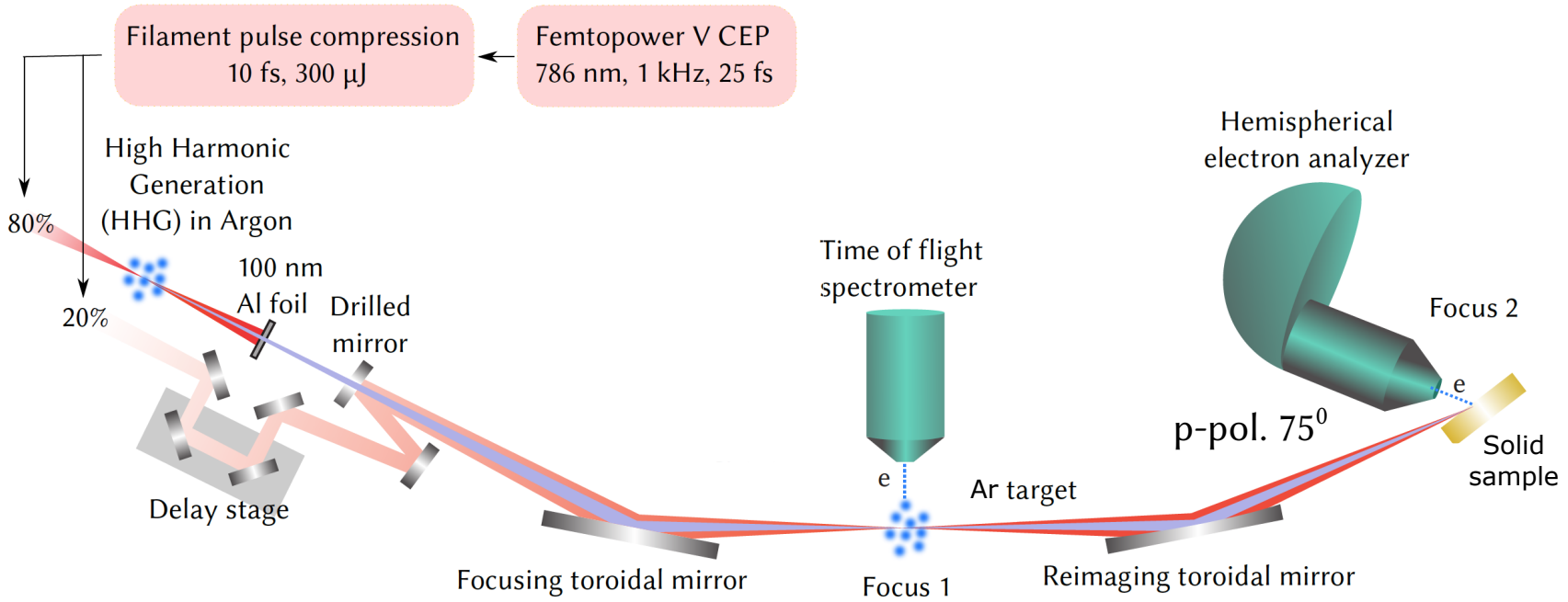
Experimental setup

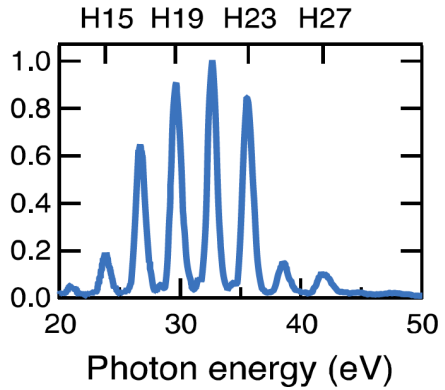


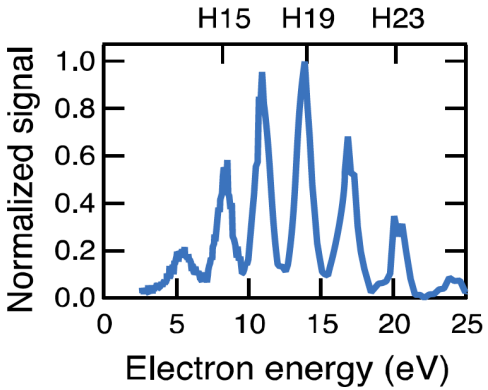


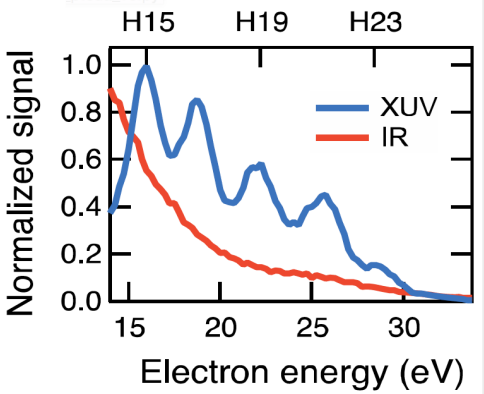
R. Locher et al., Rev. of Sci. Instr. 85, 013113 (2014)

Experimental setup









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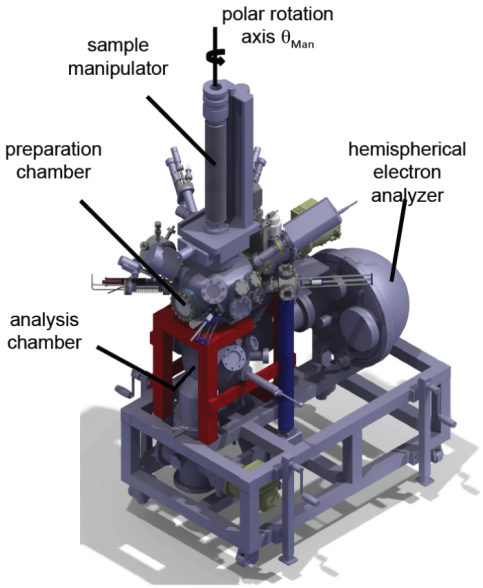
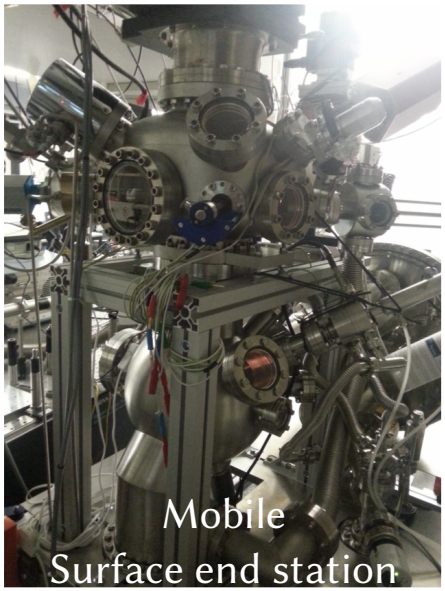
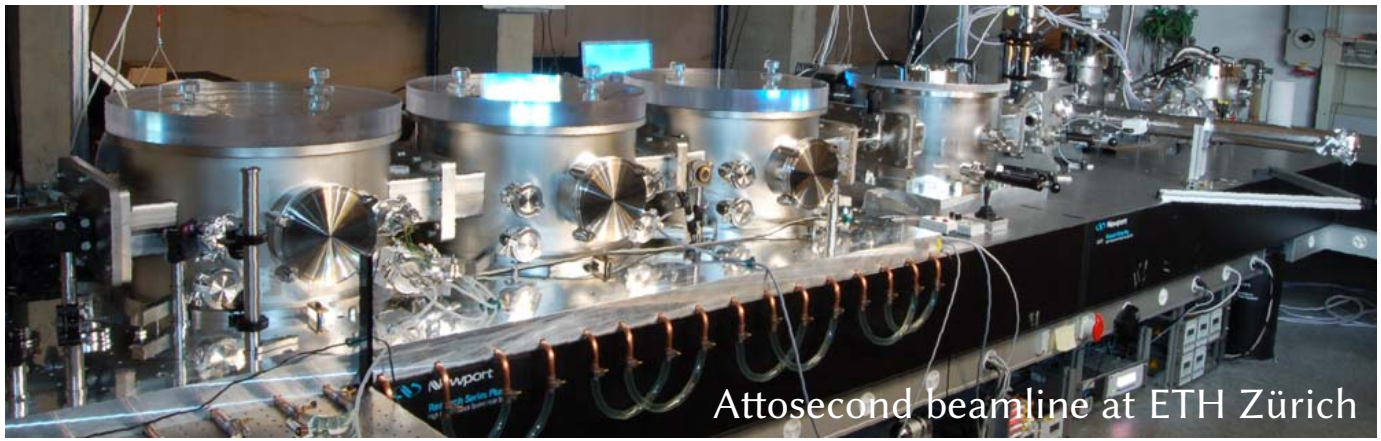
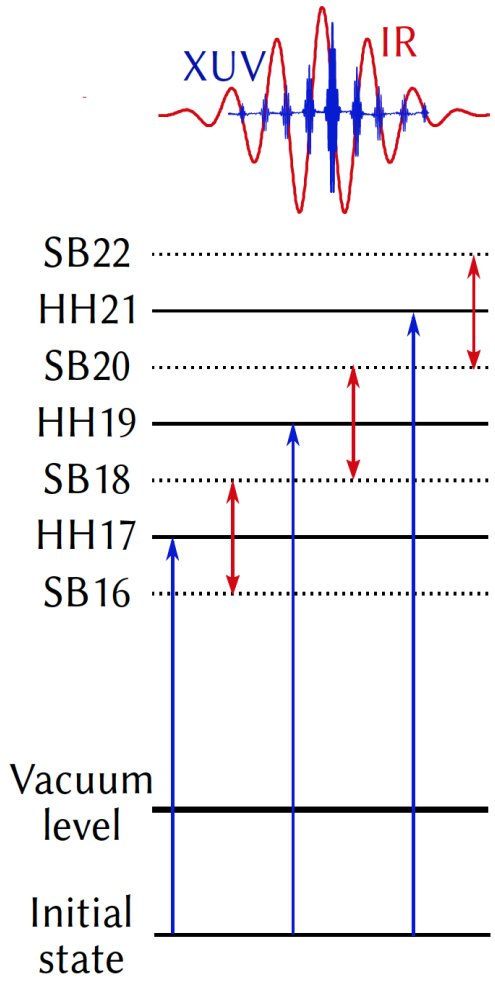




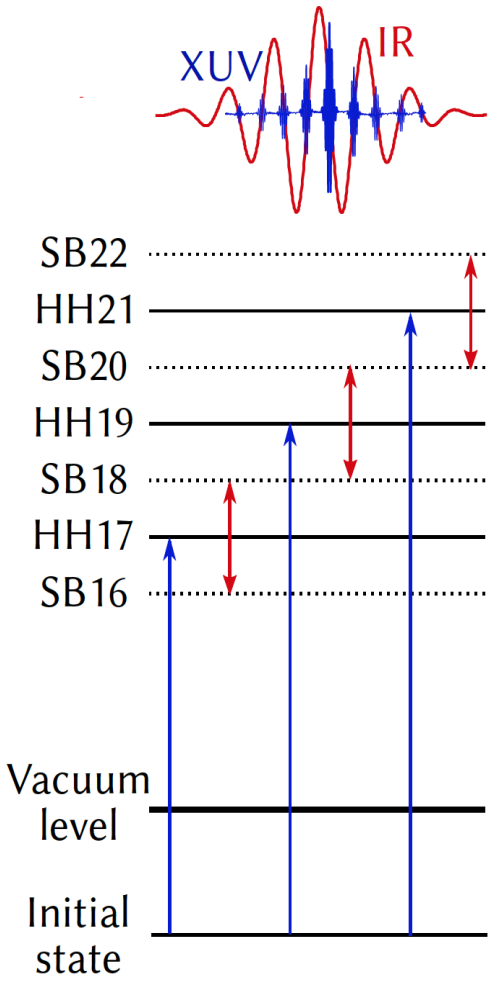


RABBITT * Reconstruction of Attosecond Beating By Interference of Two photon Transition

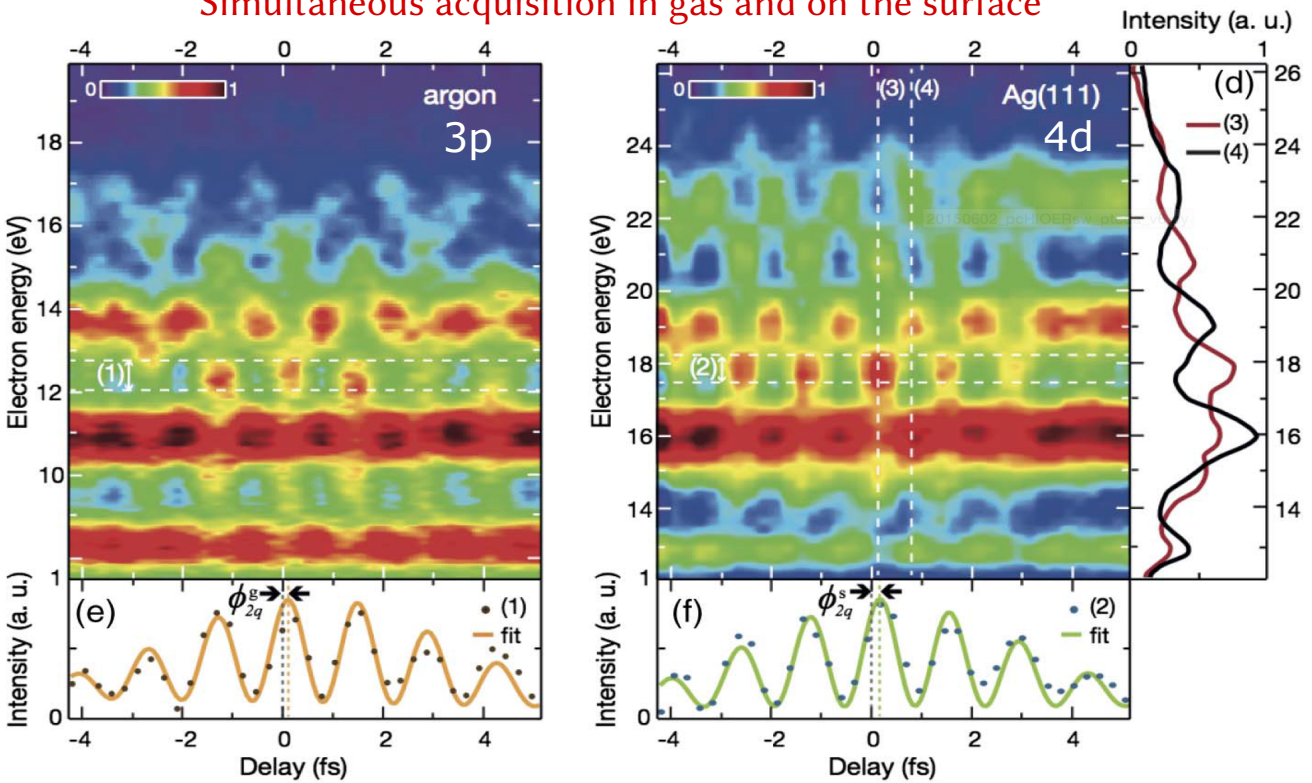


R. Locher and L. Castiglioni et al, Optica 2, 405-410 (2015)

RABBITT * Reconstruction of Attosecond Beating By Interference of Two photon Transition



Simultaneous acquisition in gas and on the surface



Integration over the energy range of SB reveals oscillation

$$SB_{2q} = A(t)cos(2\omega_{IR}t + \Phi_{2q})$$

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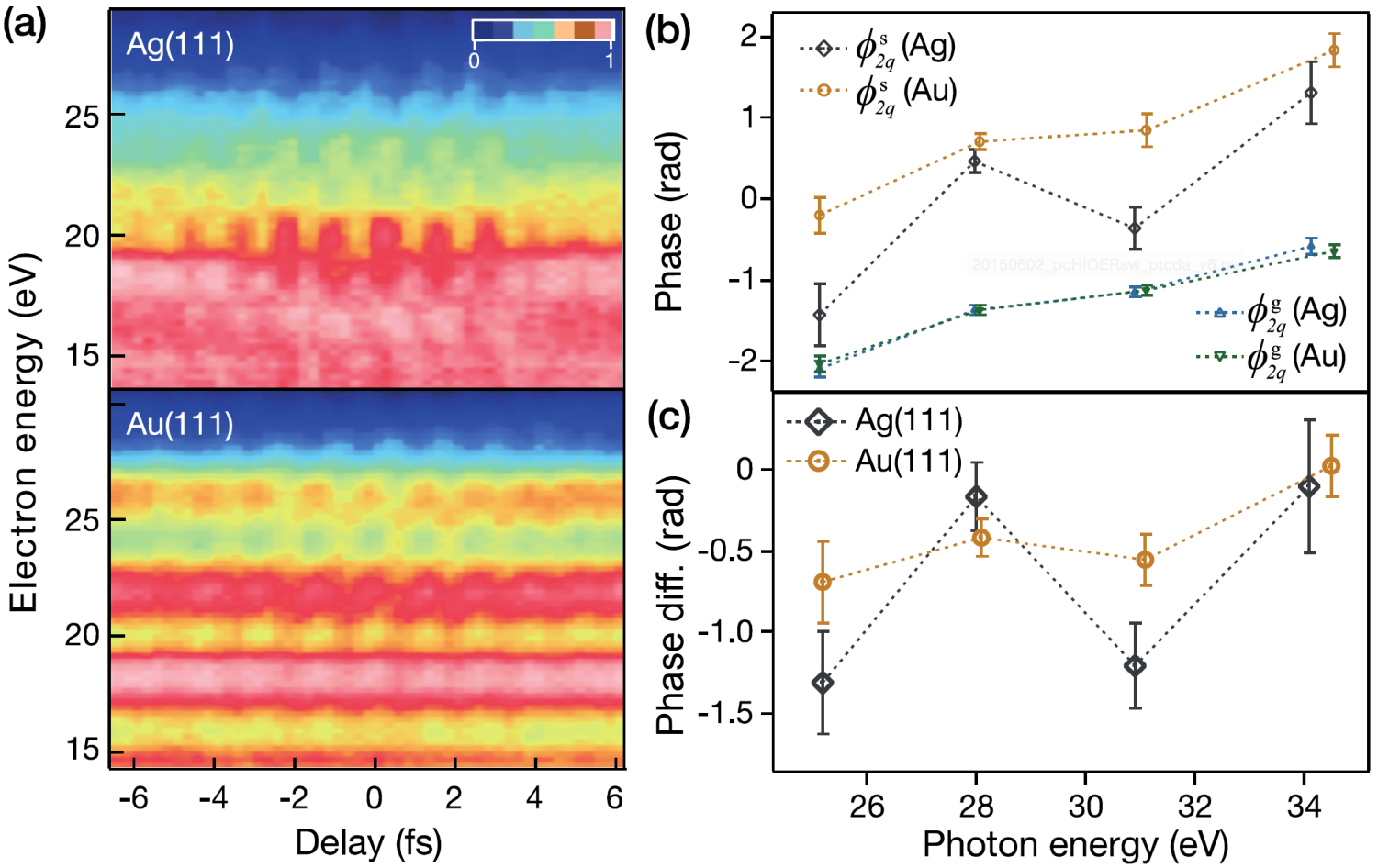
RABBITT on Ag(111) and Au(111)

Typical RABBITT traces from Ag(111) and Au(111)

Extracted raw phases from respective surface and argon traces



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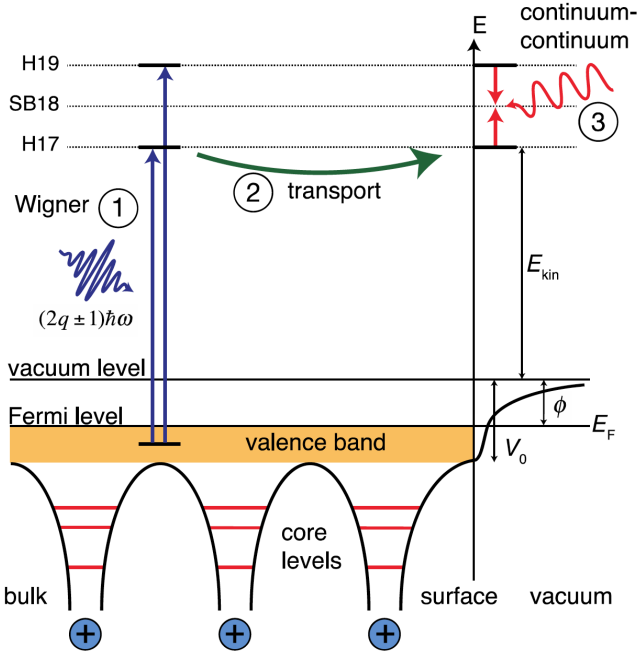
How to obtain surface specific photoemission delay



Surface specific photoemission delay

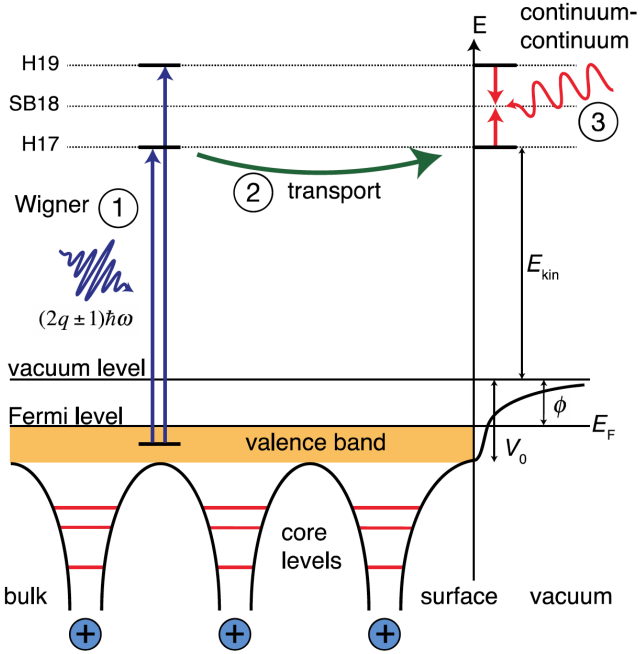
Continuum-continuum IR induced (solid)

$$\tau^s_{2q} = \tau^s_{\lambda,2q} + \tau^s_{cc,2q} + \tau^s_{trans}$$

Wigner delay (solid)

Transport within the solid

How to obtain surface specific photoemission delay



Surface specific photoemission delay — Continuum-continuum IR induced (solid)

$$\tau^s_{2q} = \tau^s_{\lambda,2q} + \tau^s_{cc,2q} + \tau^s_{trans}$$

Wigner delay (solid) — Transport within the solid

and since

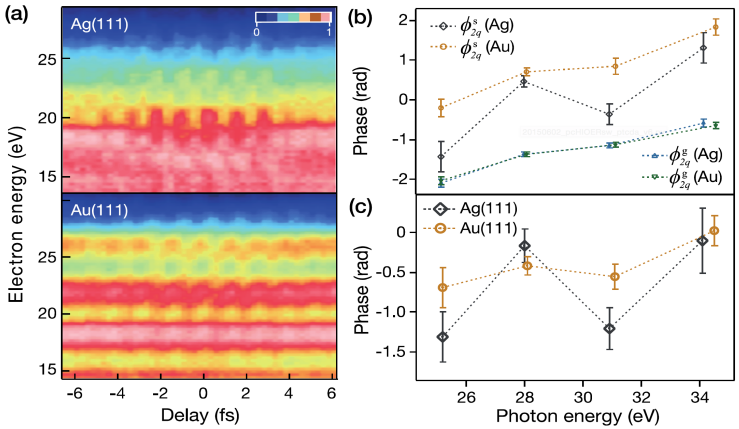
$$\begin{aligned}\phi_{2q} &= (\theta_{2q+1} - \theta_{2q-1}) + (\varphi_{2q+1} - \varphi_{2q-1}) + (\varphi_{cc,2q+1} - \varphi_{cc,2q-1}) - 2\phi_0 \\ &= \Delta\theta_{2q} + \Delta\varphi_{2q} + \Delta\varphi_{cc,2q} - 2\phi_0 \\ &= 2\omega \cdot (\tau_{GD,2q} + \tau_\lambda + \tau_{cc} - \tau_0).\end{aligned}$$

Surface specific photoemission delay — Experimental phases — Continuum-continuum IR induced (gas) — IR transient grating

$$\tau^s_{2q} = \frac{\Phi^s_{2q} - \Phi^g_{2q}}{2\omega_{IR}} + \tau^g_{\lambda,2q} + \tau^g_{cc,2q} - \tau_{prop} + \tau_{refl}$$

Wigner delay (gas) — Toroidal mirror reflection, Gouy phase

How to obtain surface specific photoemission delay



Surface specific photoemission delay — Experimental phases — Continuum-continuum IR induced (gas) — IR transient grating

$$\tau_{2q}^{s} = \frac{\Phi_{2q}^{s} - \Phi_{2q}^{g}}{2\omega_{IR}} + \tau_{\lambda,2q}^{g} + \tau_{cc,2q}^{g} - \tau_{prop} + \tau_{refl}$$

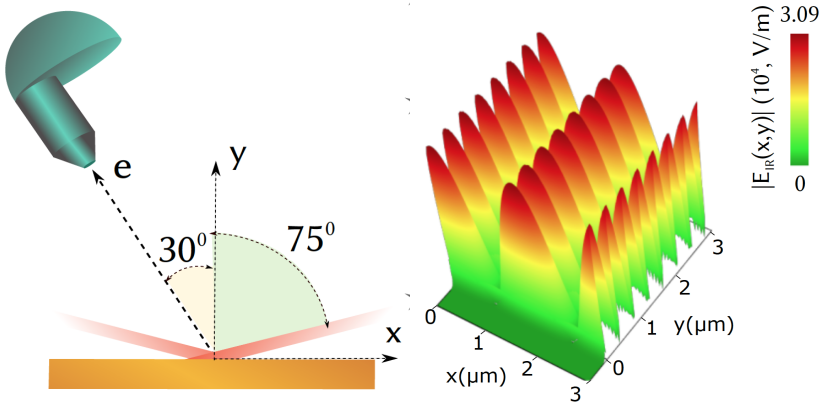
Wigner delay (gas) — Toroidal mirror reflection, Gouy phase

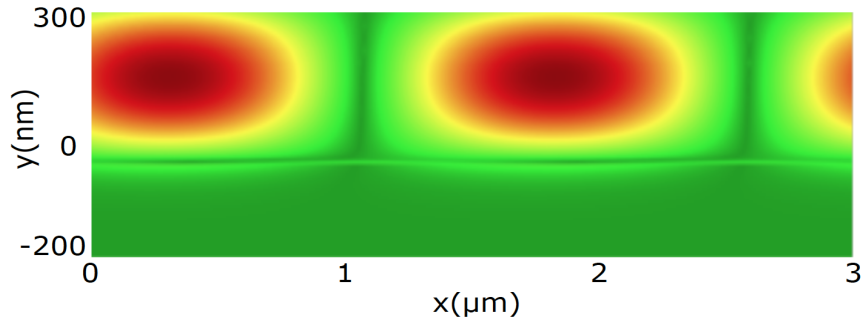
Wigner and continuum-continuum delays for Ar are taken from literature (J.M. Dahlström et al. Chem. Phys. 414, 53-64 (2013), J. Maurittson et al. Phys. Rev. A 72, 013401 (2005).)

Propagation phase is determined experimentally by performing a simultaneous RABBITT measurement with Ar in both foci: ϕ = -0.62 ± 0.14 rad, corresponds to -136±30 as

There is an additional contribution from IR transient grating due to the interference between incident and reflected IR beams.

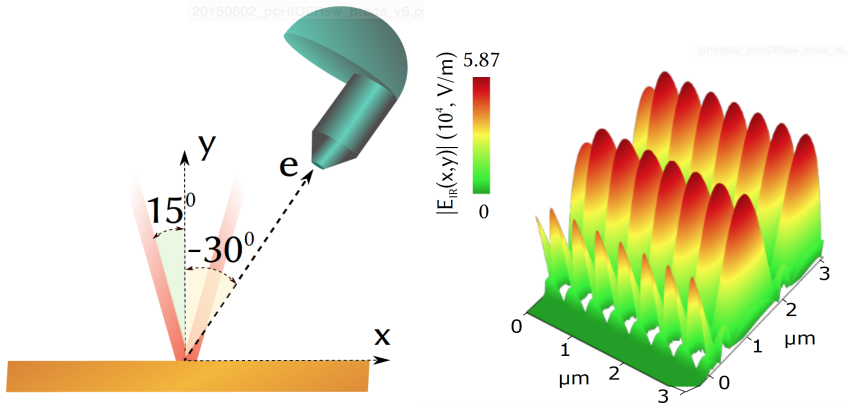
Effect of IR transient grating

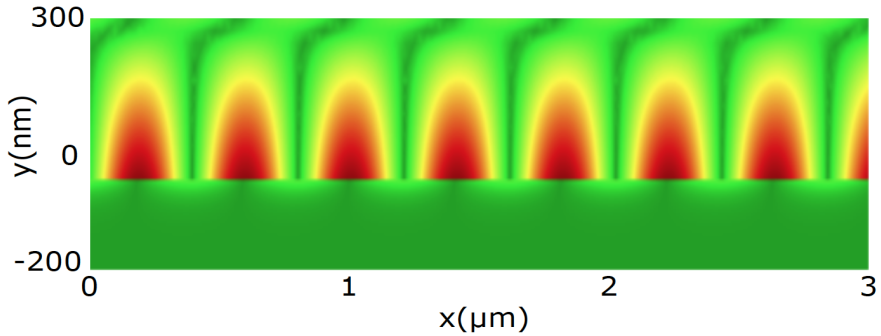




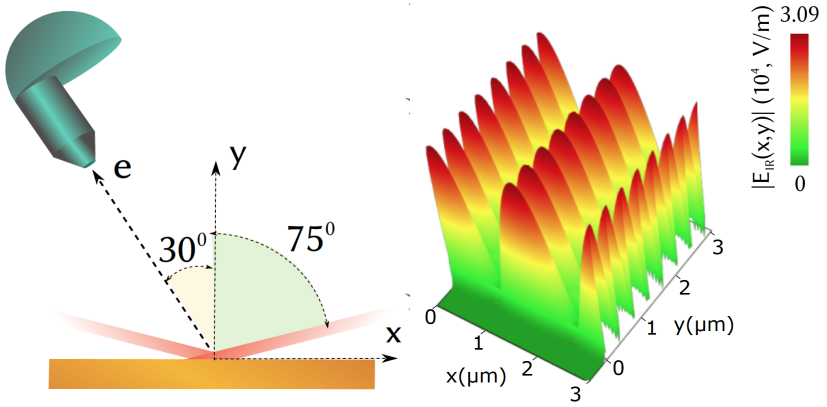
Electric field inside the solid is a strongly damped evanescent wave

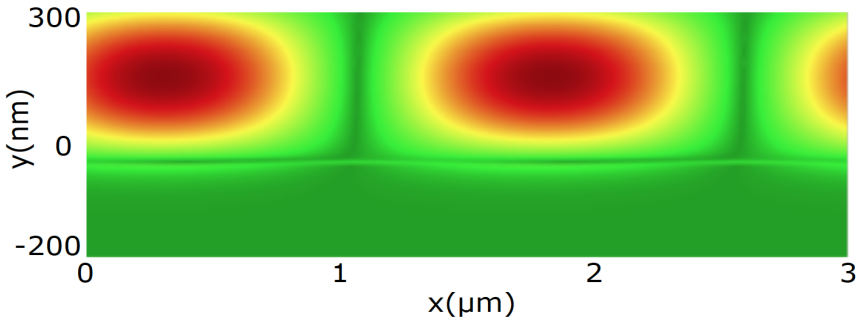
Interference between incident and reflected IR beams outside the sample leads to a formation of an angular dependent transient grating





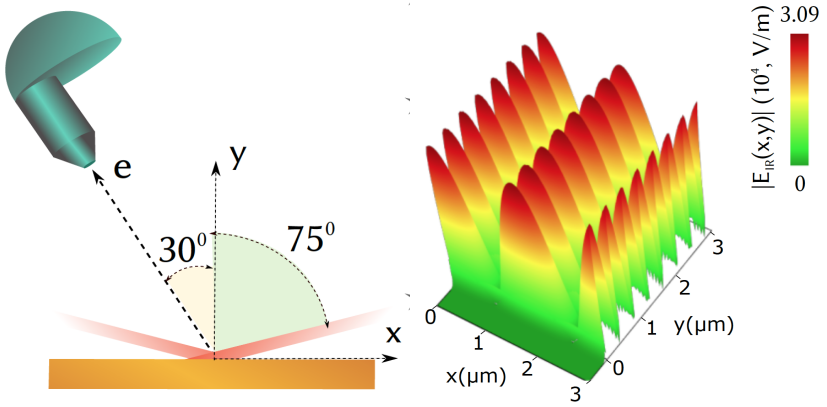
Effect of IR transient grating

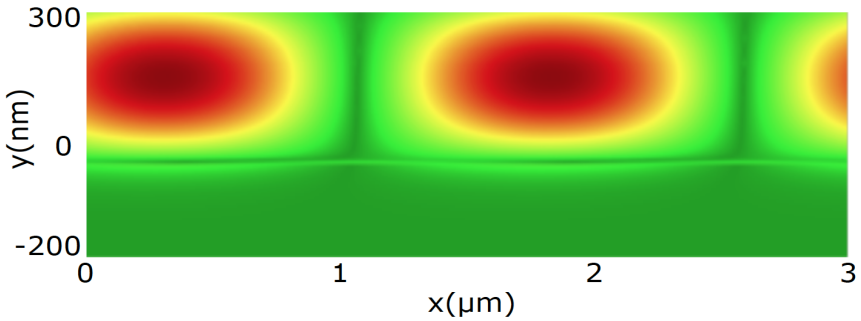




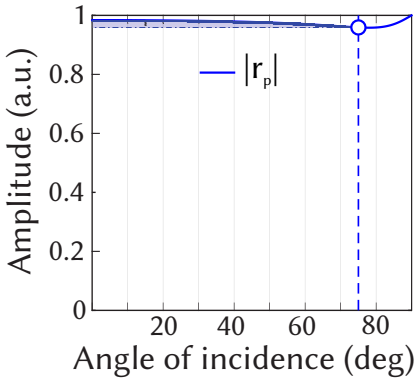
How do we obtain a phase induced by the transient grating?

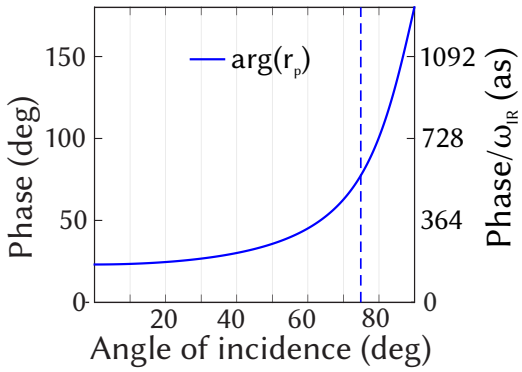
Effect of IR transient grating



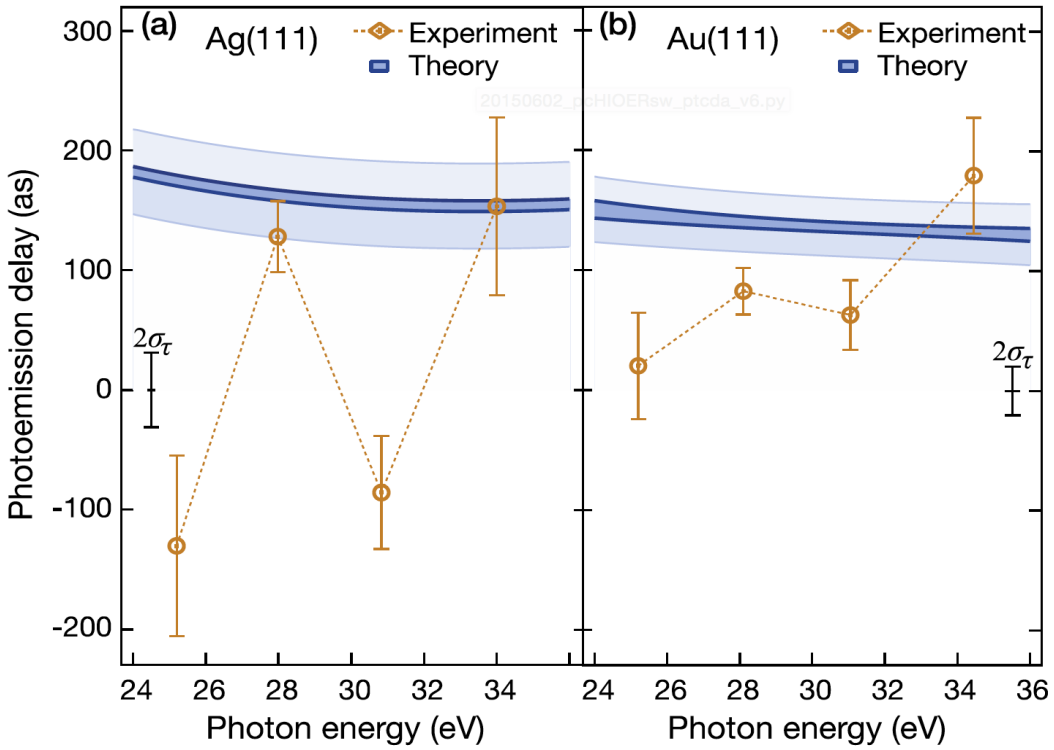


Fresnel complex reflection coefficient





Experimentally determined delays



Good agreement for 28 eV and 34 eV for Ag

Delay is dominated by transport due to interband transitions
in case of available bulk final states

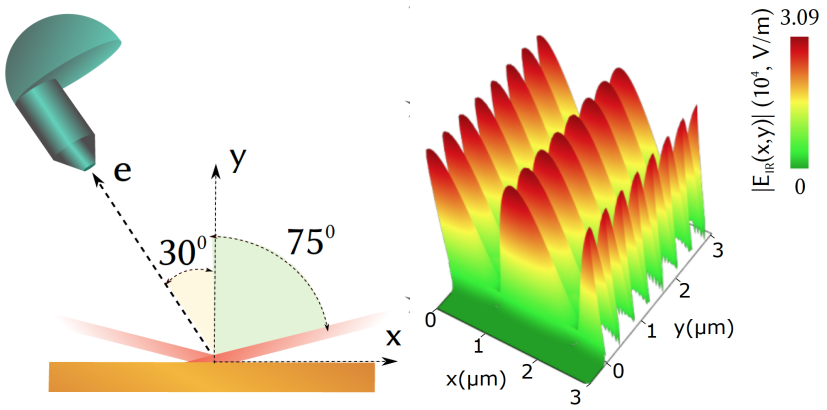
In the absence of such resonances, the delay is dominated by surface emission: 25 eV ans 31 eV for Ag

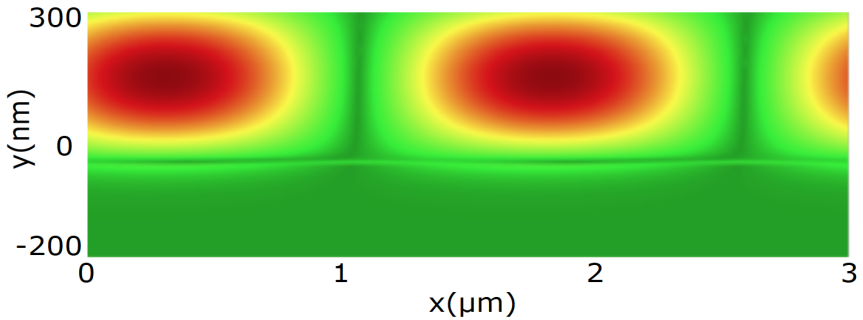
R. Locher and L. Castiglioni et al, Optica 2, 405-410 (2015)

Summary and Outlook

- ✓ RABBITT technique extended to a solid-state system
- ✓ Energy-dependent surface specific photoemission delays from Ag(111) and Au(111) in a good agreement with theory

Outlook: To the question of the IR transient grating





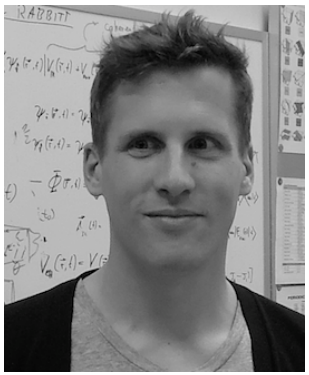
Accurate determination of photoemission delays requires through disentangling of all contributions.

What is the true contribution of the IR transient grating induced phase on the photoemission delays?

Can we apply Fresnel equations for the description of the electromagnetic fields at atomic length- and attosecond time scales relevant to the RABBITT process?

Thank you for your attention!

Acknowledgements


Luca Castiglioni


Michael Greif


Lamia Kasmi


Matteo Lucchini


Jürg Osterwalder


Lukas Gallmann


Matthias Hengsberger


Ursula Keller

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