Interferometric photoemission from surfaces

Recall our derivation of the probability for interferometric electron emission from a discrete state $|i\rangle$ by an attosecond pulse train with a delayed (or advanced) IR pulse:

$$P_i(\vec{k}, \tau) = \Big| \sum_j E_{xuv,j} \, e^{i[\phi_j^{xuv} - \omega_j \tau]}$$

$$\times \sum_n \frac{(-)^{|n|}}{2^{|n|}\,|n|!} \left(k \, E_{IR,0} \cos\Theta \right)^{|n|}$$

$$\times \int dt \; f_{xuv}(t+\tau) \left[f_{IR}(t) \right]^{|n|} \, |d_{\vec{k},i}| \, e^{i\varphi_j(t)}$$

$$\times \, e^{-i[\varepsilon_i + (j+n)\omega_{IR} - \varepsilon_{\vec{k}}]t} \Big|^2 ,$$

where paths (j, n) are added coherently.

For valence-band emission we generalize this result by

i) replacing $|i\rangle$ by states of Bloch momentum $\vec{k}_i$ in band m: $|m, \vec{k}_i\rangle$

ii) incoherently adding occupied initial states:

$$P(\vec{k}, \tau) = \sum_m \int d\vec{k}_i \; \rho_m(\vec{k}_i) \, f(k_i, T) \; P_{|m,\vec{k}_i}(\vec{k}, \tau)$$

$\rho_m(\vec{k}_i)$: density of states

$f(k_i, T)$: FD distribution, T: abs. temp.

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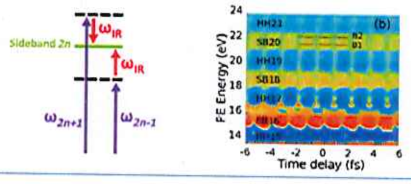
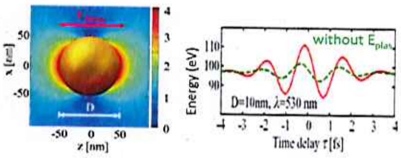
Lecture 12: Outline

1. RABBITT interferograms
- Electronic structure & dielectric response
- Examples: *Cu(100/111)*
- Final-state resonances



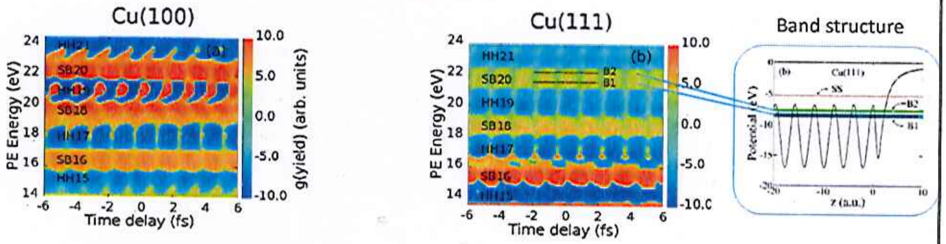
2. Plasmonic near-field imaging
- Methods: *CTMC, S-matrix*
- Examples: *Au, Ag, Cu nanospheres*
- Strong field ionization
- Streaking with a twist



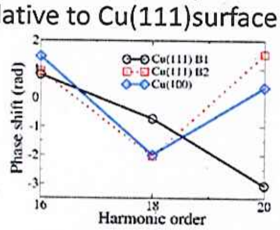
Uwe Thumm, KSU 1

1

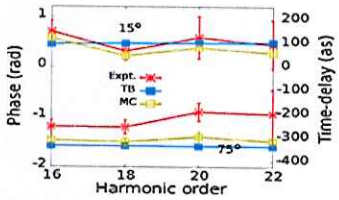
RABBITT spectra from Cu surfaces
XUV-background subtracted



Cu RABBITT phases relative to Cu(111)surface state



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



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

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

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

PQE 19 Uwe Thumm, KSU 2

2

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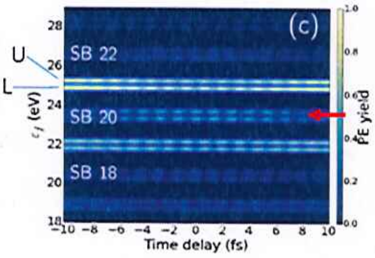
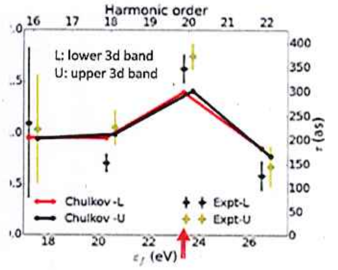
RABBITT spectra from Cu(111) surfaces

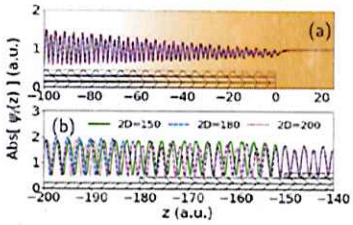
Final-state - solid interaction effects





Final-state resonance in substrate potential:

- enhances SB 20 yield
- significantly increases RABBITT phase shift

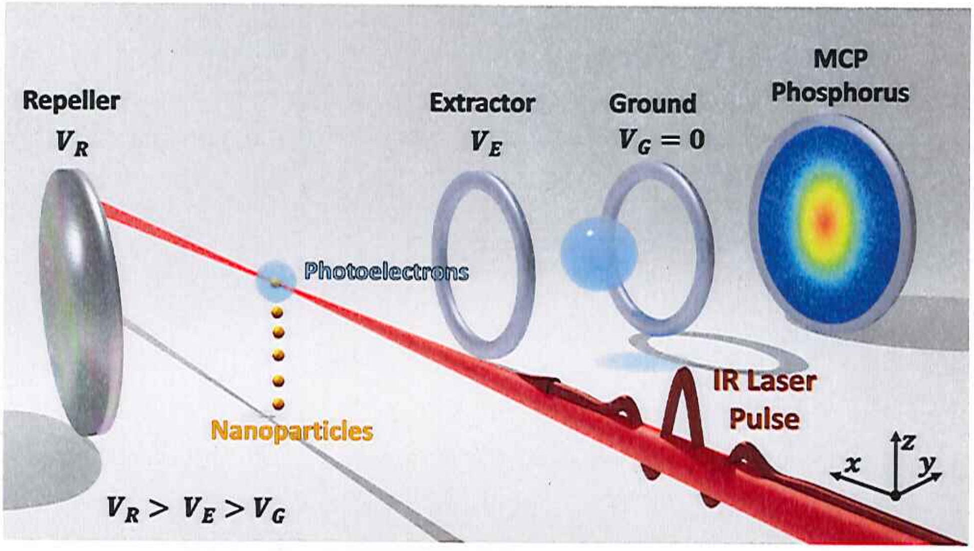


Theory: Ambrosio, U.T., *PRA* **100**, 043412 (2019)
Expt.: Kasmi,..., Gallmann, Keller, *Optica* **4**, 1492 (2017)

PQE 19 Uwe Thumm, KSU 3

3

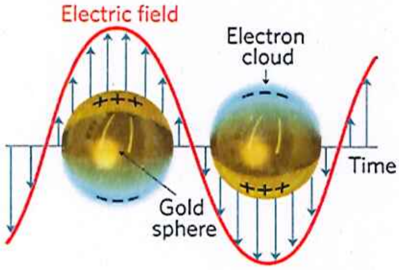
Strong-field ionization of metal nanoparticles



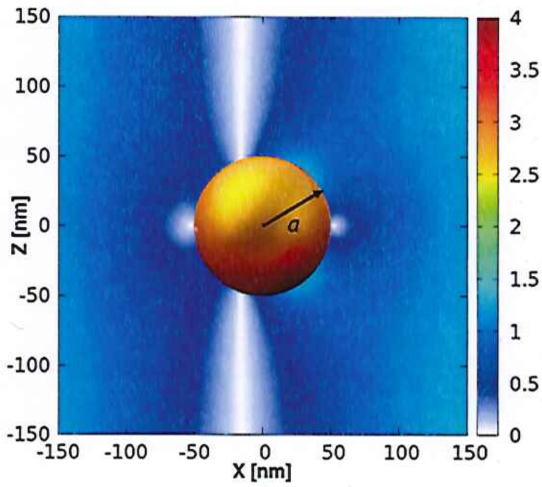
1

Induced plasmonic field

Localized surface charge oscillation (plasmon)



Juan *et. al.*, Nature Photonics **5**, 349 (2011)



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Plasmonic and total electric field

Incident field

$$\vec{E}_{inc}(t) = \sqrt{I_0}\,\exp\left[-2\ln 2\frac{(t-x/c)^2}{\tau^2}\right]\exp\left(-i\omega\left(t-\frac{x}{c}\right)\right)\hat{k}$$

Induced plasmonic field

$$\vec{E}_{pl}(\vec{r},t) = k^2\left(\hat{r}\times\vec{P}(\vec{r},t)\right)\times\hat{r}\frac{e^{ikr}}{r} + \left(3\hat{r}\left(\hat{r}\cdot\vec{P}(\vec{r},t)\right)-\vec{P}(\vec{r},t)\right)\left(\frac{1}{r^3}-\frac{ik}{r^2}\right)e^{ikr}$$

Total field

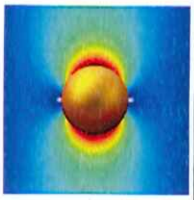
$$\vec{F}(\vec{r}_i,t) = \vec{E}_{inc}(t) + \vec{E}_{pl}(\vec{r}_i,t) - Z(t)\frac{\vec{r}_i}{|\vec{r}_i|^3}$$

residual charge

Induced polarization

$$\vec{P}(\vec{r},t) = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} d\omega e^{i\omega t}\,\vec{P}(\vec{r},\omega)$$

$$\vec{P}(\vec{r},\omega) = \epsilon_0\epsilon_m\alpha_{Mie}(\omega)\,\vec{E}_{inc}(\vec{r},\omega)$$



Polarizability

$$\alpha_{Mie}(\omega) = \frac{1-\frac{1}{10}(\epsilon(\omega)+\epsilon_m)x^2}{\left(\frac{1}{3}+\frac{\epsilon_m}{\epsilon(\omega)-\epsilon_m}\right)-\frac{1}{30}(\epsilon(\omega)+10\epsilon_m)x^2-i\frac{4\pi^2\epsilon_m^{\frac{3}{2}}}{3}\frac{V}{\lambda_0^3}}$$

$$x = \frac{2\pi a}{\lambda_0}$$

H. Kuwata, *et al.*, Appl. Phys. Lett. **83**, 4625 (2003)

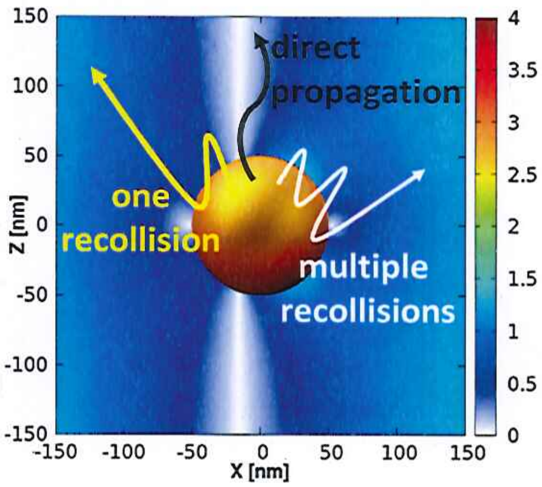
E. D. Palik, (Ed.) *Handbook of optical constants of solids*, Vol. **3** (1998)

3

Photoelectron momentum distribution
Two-step theory

1 Photoelectron emission from the surface

2 Photoelectron propagation to the detector



4

1 Photoelectron emission from the surface

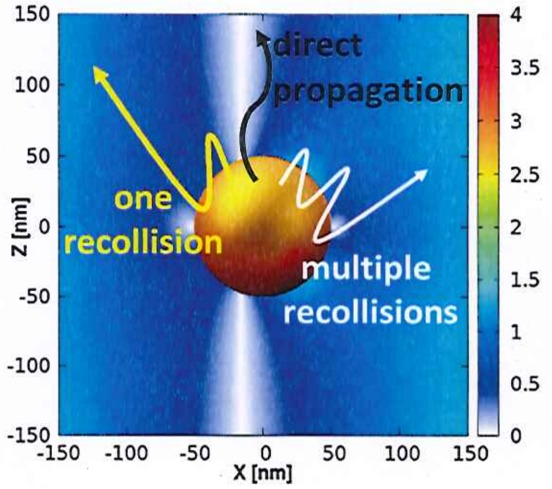
Photoelectron yield: N_e

↑

Photoelectron current density: $J_r(\vec{r_s}, t)$

↑

Tunneling ionization rate: $W(p_r, F)$



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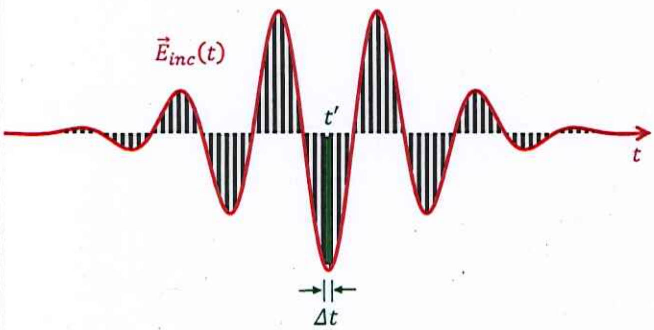
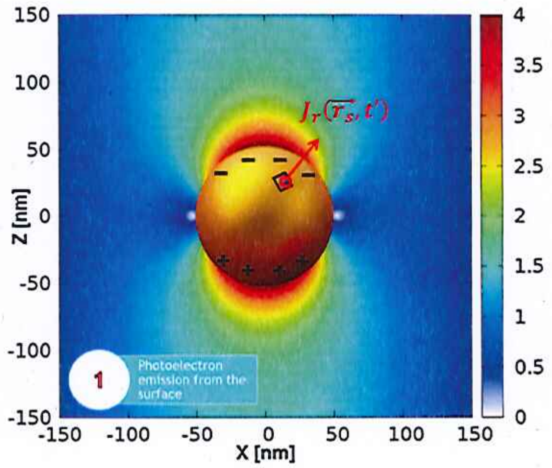
Photoelectron yield



$$\Delta N_e(t') = \Delta t \int_0^{2\pi} d\phi \int_0^{\pi} d\theta \; a^2 \; \sin\theta \; J(\vec{r_s}, t')$$



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Photoelectron current density

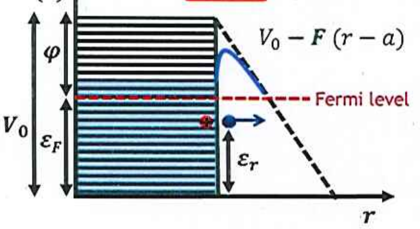
$$J_r(\vec{r_s},t) = \int_{-\infty}^{\infty} d\vec{p}\, g(\vec{p})\; f_{FD}(\vec{p})\; W(p_r,F)\, p_r$$

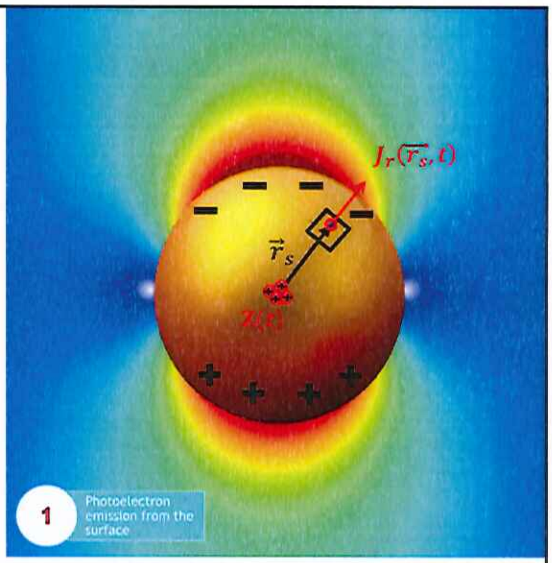
Modified Fowler-Nordheim tunneling ionization rate

$$W(\varepsilon_r,F) \cong 4\,\frac{\sqrt{\varepsilon_r(V_0-\varepsilon_r)}}{V_0}\; exp\left[-\frac{4\sqrt{2}}{3F}\,v(f)(V_0-\varepsilon_r)^{\frac{3}{2}}\right]$$

Barrier shape correction

$$N_e(t) = \Delta t \int_0^{2\pi} d\phi \int_0^{\pi} d\theta\; a^2\; \sin\theta\; J(\vec{r}_s,t)$$





2 Propagation to the detector

Newton's equations

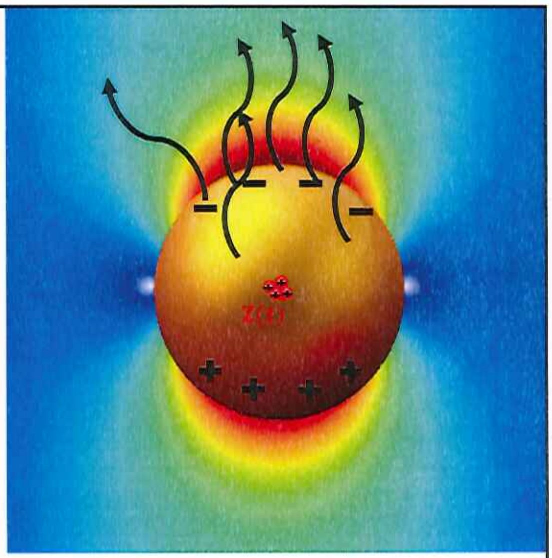
$$\frac{d\vec{v}_i}{dt} = -\vec{E}_{inc}(t) - \vec{E}_{pl}(\vec{r}_i,t) - Z(t)\frac{\vec{r}_i}{|r_i|^3} + \frac{1}{2}\sum_{j\neq i}^{Z(t)} \frac{\vec{r}_i-\vec{r}_j}{|\vec{r}_i-\vec{r}_j|^3}$$

$$\frac{d\vec{r}_i}{dt} = \vec{v}_i$$

electron - residual charge interaction

electronic interaction

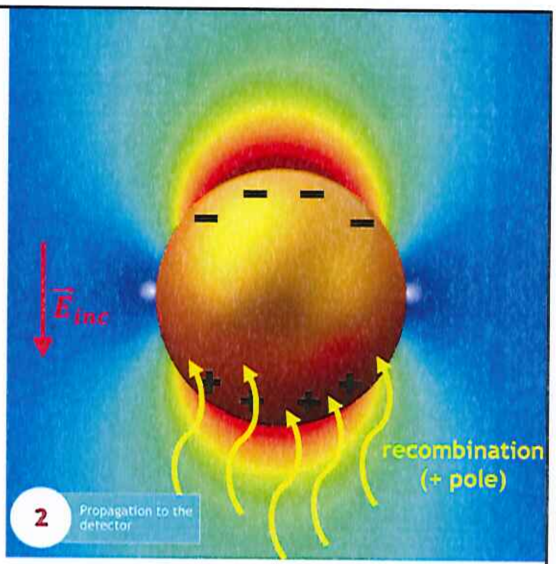
$$Z(t) = \int_0^t dt' N_e(t')$$



Monte Carlo sampling

See: E. Saydanzad, J. Li, U. Thumm, Phys. Rev. A **95**, 053406 (2017)

Recombination at + pole



9

Recombination & rescattering (- pole)

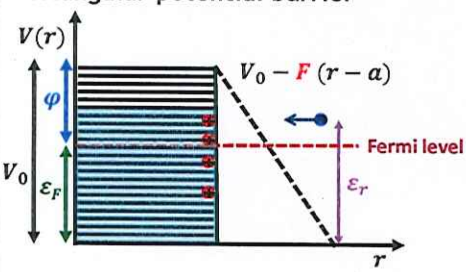
Inverse tunneling rate

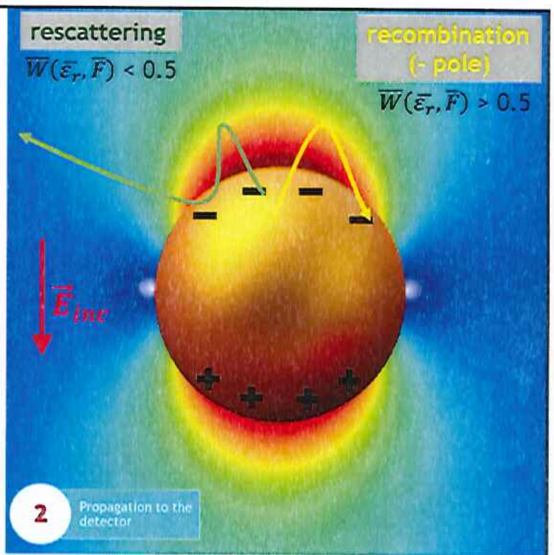
P. Zhang and T. Pan, AIP Advances 7, 065307 (2017)

$$\overline{W}(\bar{\varepsilon}_r,\bar{F}) = 4\frac{\sqrt{\bar{\varepsilon}_r(1+\bar{\varepsilon}_F-\bar{\varepsilon}_r)}}{1+\bar{\varepsilon}_F}\, exp\left[-\frac{4\sqrt{2}}{3\bar{F}}(1+\bar{\varepsilon}_F-\bar{\varepsilon}_r)^{\frac{3}{2}}\right]$$

$$\bar{\varepsilon}_F = \frac{\varepsilon_F}{\varphi} \qquad \bar{\varepsilon}_r = \frac{\varepsilon_r}{\varphi} \qquad \bar{F} = \frac{F}{\sqrt{2}\varphi^{\frac{3}{2}}}$$

Triangular potential barrier





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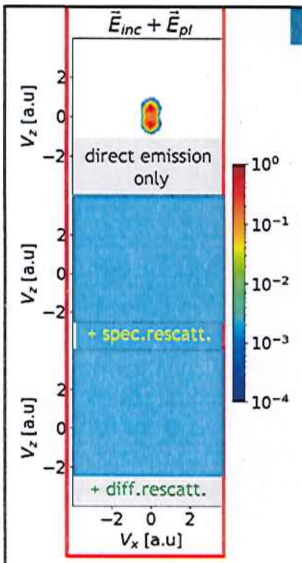
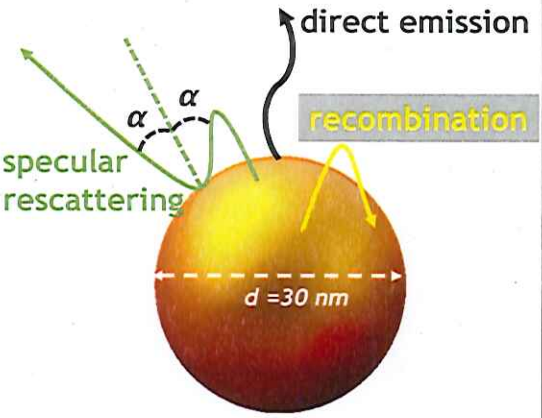
Recombination & rescattering

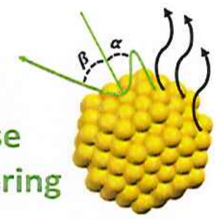


Effect on photoelectron momentum distribution

$$\frac{d\vec{v}_i}{dt} = -\vec{E}_{inc}(t) - \vec{E}_{pl}(\vec{r}_i, t)$$



diffuse rescattering



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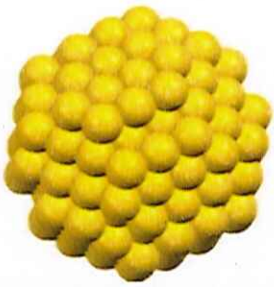
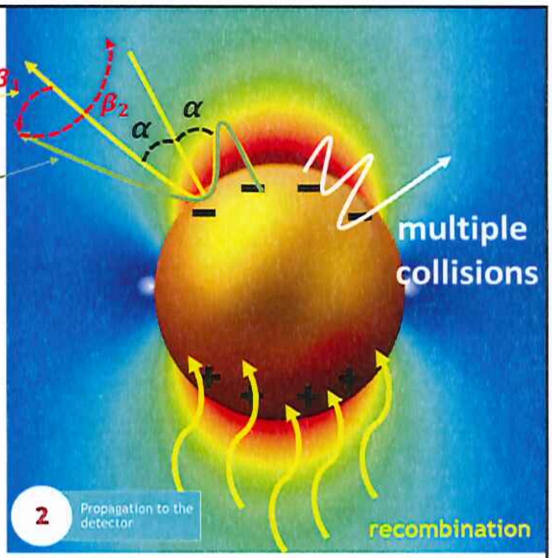
Recombination & rescattering

Specular rescattering

Diffuse rescattering





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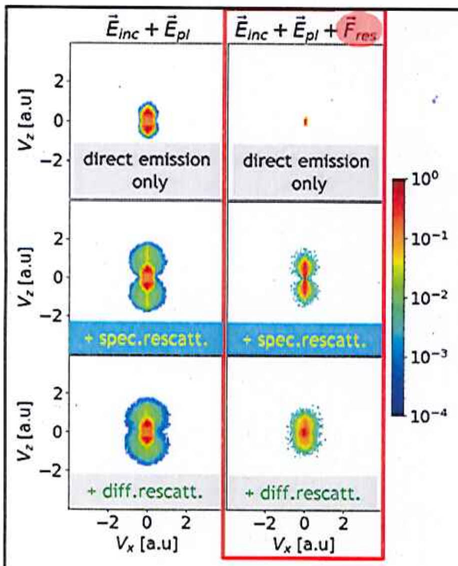
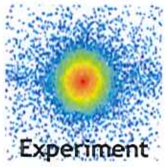


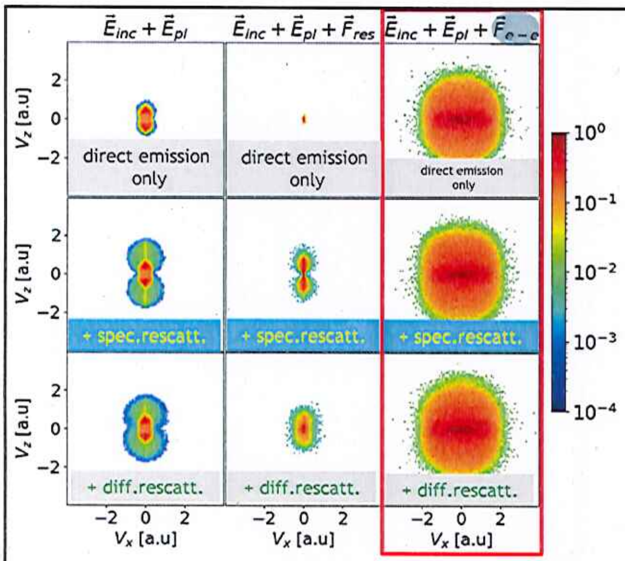
Recombination & rescattering

Added effect of:
residual charges

$$\frac{d\vec{v}_i}{dt} = -\vec{E}_{inc}(t) - \vec{E}_{pl}(\vec{r}_i, t) - \boxed{Z(t)\frac{\vec{r}_i}{|\vec{r}_i|^3}}$$

$$= \vec{F}_{res}$$



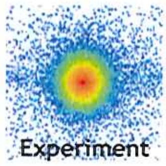


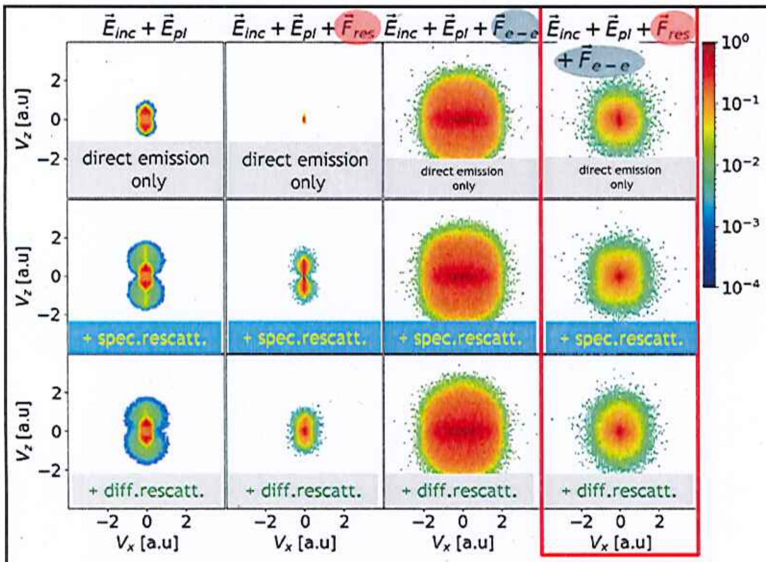
Recombination & rescattering

Added effect of
photoelectron correlation

$$\frac{d\vec{v}_i}{dt} = -\vec{E}_{inc}(t) - \vec{E}_{pl}(\vec{r}_i, t) + \boxed{\frac{1}{2}\sum_{j \neq i}^{Z} \frac{\vec{r}_i - \vec{r}_j}{|\vec{r}_i - \vec{r}_j|^3}}$$

$$= \vec{F}_{e-e}$$





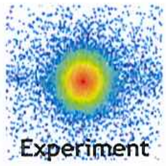
Recombination & rescattering

Added effects of residual charges and photoelectron correlation

$$\frac{d\vec{v}_i}{dt} = -\vec{E}_{inc}(t) - \vec{E}_{pl}(\vec{r}_i, t) - Z(t)\frac{\vec{r}_i}{|\vec{r}_i|^3} + \frac{1}{2}\sum_{j\neq i}^{Z}\frac{\vec{r}_i - \vec{r}_j}{|\vec{r}_i - \vec{r}_j|^3}$$

$= \vec{F}_{res}$

$= \vec{F}_{e-e}$



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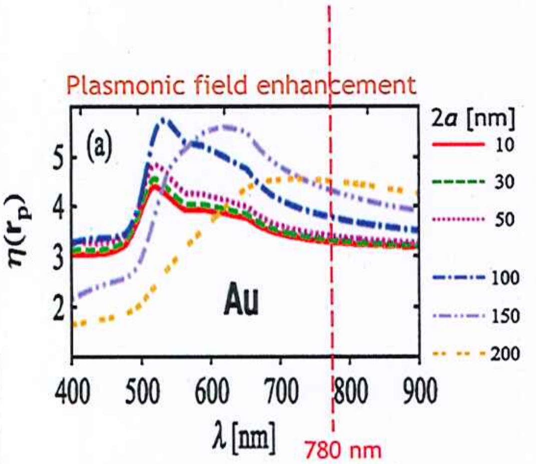


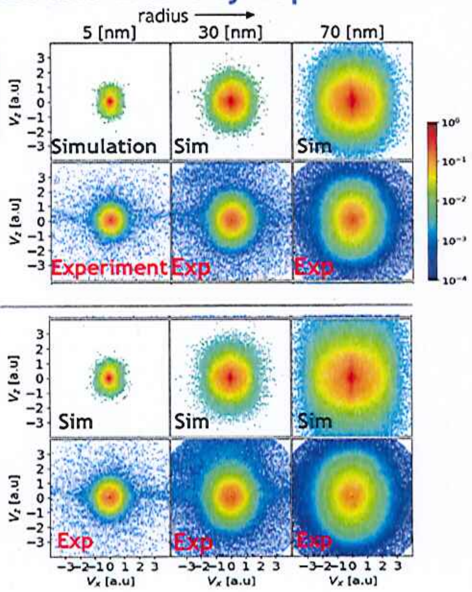
Li, Saydanzad, Thumm, Phys. Rev. A **95**, 043423 (2017)

Photoelectron momentum distributions

Size and intensity dependence

I_0=8x10^{12} W/cm^2
τ = 25 fs
λ = 780 nm

$1.5\, I_0$



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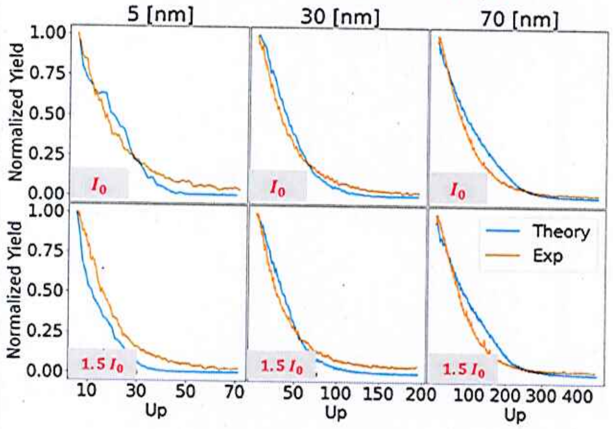
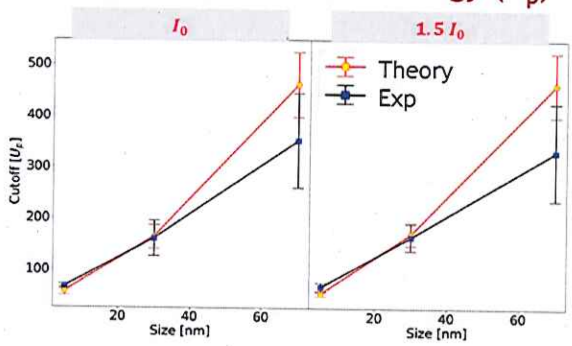
Photoelectron momentum distributions
Size and intensity dependence

Photoelectron yield (U_p)



Photoelectron cutoff energy (U_p)



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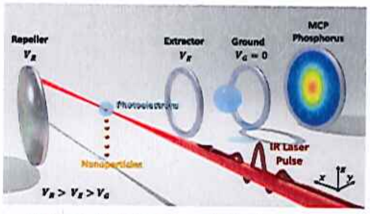
Strong-field ionization of metal nanoparticles

Erfan Saydanzad



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Adam Summers (ICFO, KSU)
Philipp Rupp (LMU)
Christopher M Sorensen (KSU)
Daniel Rolles (KSU)
Matthias F Kling (LMU)
Carlos Trallero-Herrero (UConn)
Artem Rudenko (KSU)

Jeffrey Powell

E. Saydanzad, J. Li, U. T. + exp. collaborators, in progress (2022)

E. Saydanzad, J. Li, U. Thumm, Phys. Rev. A **98**, 063422 (2018)
J. Li, E. Saydanzad, U.Thumm, Phys. Rev. Lett **120**, 223903 (2018)
E. Saydanzad, J. Li, U. Thumm, Phys. Rev. A **95**, 053406 (2017)
J. Li, E. Saydanzad, U. Thumm, Phys. Rev. A **95**, 043423 (2017)
J. Li, E. Saydanzad, U. Thumm, Phys. Rev. A **94**, 051401(R) (2016)

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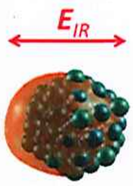
Streaking with a twist

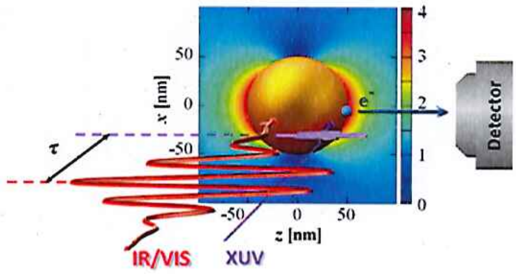
Reconstruction of nanoplasmonic near fields

Probing collective electronic dynamics in nano-particles

Plasmonic near field emhancement



Imaging plasmonic fields near Au nano-spheres in streaked photoelectron spectra



Leone, U.T. *et al.*, *Nat. Photon.* **8**,162 (2014)

QM model: Li, Saydanzad, U.T., *PRA* **95**, 043423 (2017)

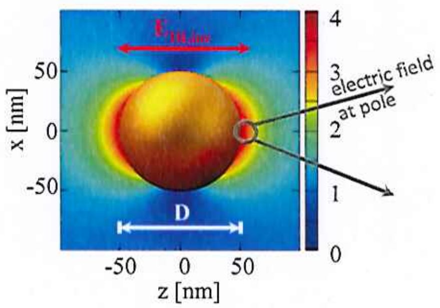
CTMC model: Saydanzad, Li, U.T., *PRA* **95**, 053406 (2017)

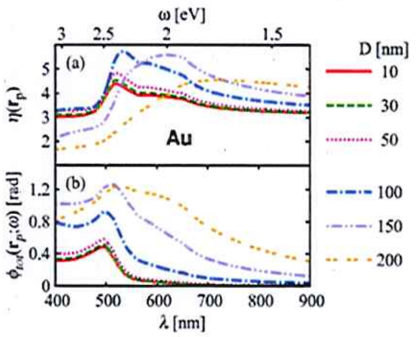
QM model: Li, Saydanzad, U.T., *PRL* **120**, 223903 (2018)

1

Plasmonic field enhancement η and phase shift ϕ

$$E_{IR,\,tot} = E_{IR,\,inc} + E_{plas}\,(D, \omega)$$





Plasmonic near-field calculation

- Mie, *Ann. Phys.* **25**, 377 (1908)
- **Dielectric response** ε(ω) from exp. data: Palik & Hunter (1985)

QM model: Li, Saydanzad, U. T., *PRA* **94**, 0514101(R) (2016)

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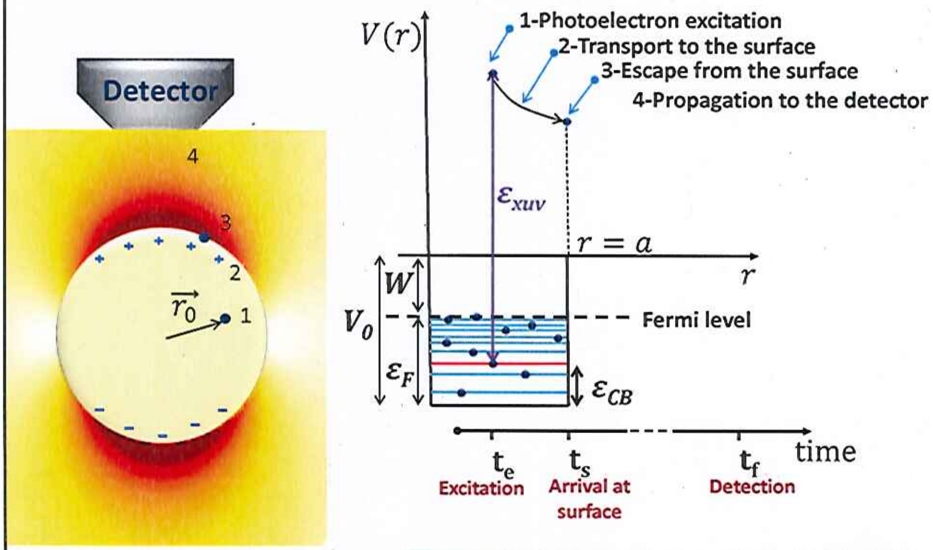
(80)

Probing collective electronic dynamics in nano-particles with streaked photoemission



CTMC model: Saydanzad, Li, U.T., *PRA* **95**, 053406 (2017)
QM model: Li, Saydanzad , U.T., *PRL* **120**, 223903 (2018)

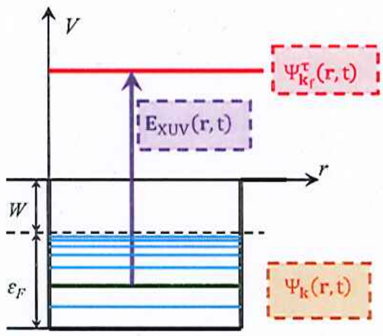
Missouri S&T Oct. 4, 2018

3

Numerical model

$$T_k(k_f,\tau) = -i\int dt \langle \Psi^{\tau}_{k_f}(r,t)|A_{XUV}(r,t)\cdot p|\Psi_k(r,t)\rangle$$

$$P(\varepsilon_f,\tau) = \sum_{k\in occ} |T_k(k_f,\tau)|^2$$



Effective potential
(sph. Square well, DFT, etc)

Initial state $\Psi_k(r,t)$
- Bound states in Au conduction band below ε_{Fermi}

Gaussian XUV pulse $E_{XUV}(r,t)$
- Au nanosphere considered transparent XUV pulse

Final state $\Psi_{k_f}(r,t)$
- Streaking-field dressed "generalized" Volkov state
- Plasmonically enhanced streaking field enters Volkov phase.

$$\phi_V = \frac{1}{2}\int_t^{\infty} dt\left(k_f + \int_t^{\infty} dt\, E_{IR,tot}(r,t)\right)^2$$

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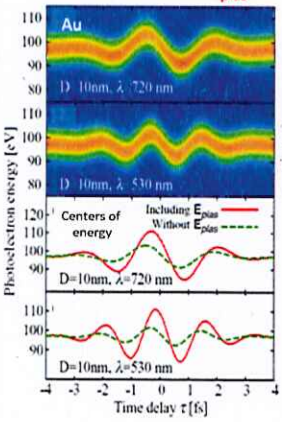
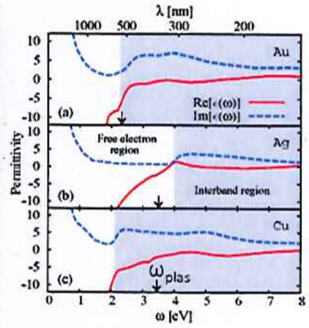
Retrieving plasmonic enhancements and phase shifts from streaked photoelectron spectra

$$E_{IR,\,tot} = E_{IR,\,inc} + E_{plas}\,(D,\,\omega)$$

streaked spectra with/without E_{plas}

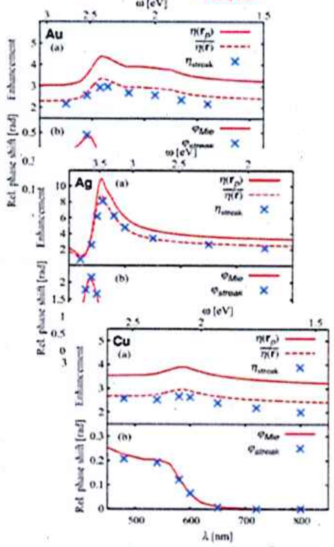


permittivity
from Palik & Hunter (1985)



max. polarizability:
$\mathrm{Re}[\varepsilon(\omega_{plas})] = -2$

x: retrieved $E_{IR,\,tot}$



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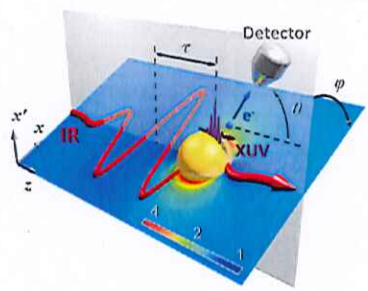
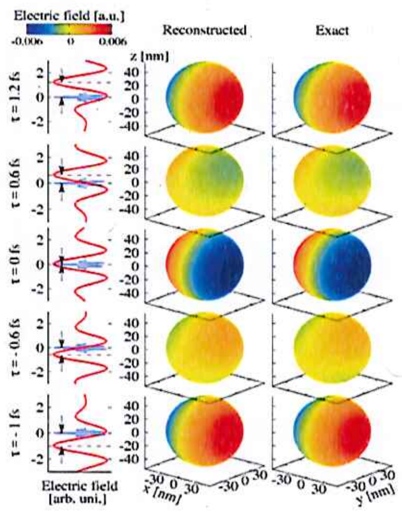
4D-streaking imaging of plasmonic fields





Reconstruction algorithm

$$E_{IR,\,tot}(\theta,\varphi,\tau) = \frac{\partial}{\partial\tau}\frac{\overline{\varepsilon_f(\tau;\theta,\varphi)} - \omega_X - (3/5)\sigma_c - V_0}{\sqrt{2\,\overline{\varepsilon_f(\tau;\theta,\varphi)}}}$$

$\overline{\varepsilon_f(\tau;\theta,\varphi)}$: Photoelectron spectral center of energy

ω_X: XUV central photon energy (50 eV)

σ_c: Conduction-band width (8 eV)

V_0: Conduction-band model potential depth (-13.1 eV)

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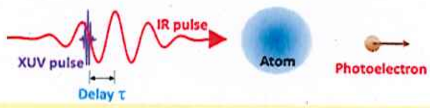
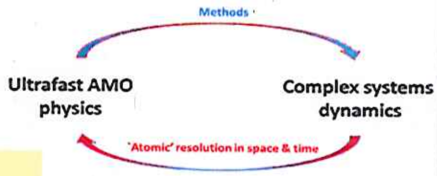
Challenges & opportunities



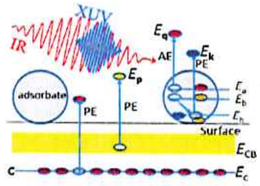


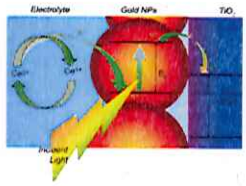
All targets
- Interpretation of streaking time delays & RABBITT phases

Surfaces/nanoparticles
Dependence of photoemission and time delay on
- electron propagation/dispersion
- accurate final state modeling
- IR /XUV skin depth, surface charges, ...

Imaging plasmonic near-fields:
- exp. validation of spatio-temporal near-field imaging
- plasmon response / collective modes

Field-dressed band-structure
- atomic resolution in space & time
- towards time-resolved ARPES....





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