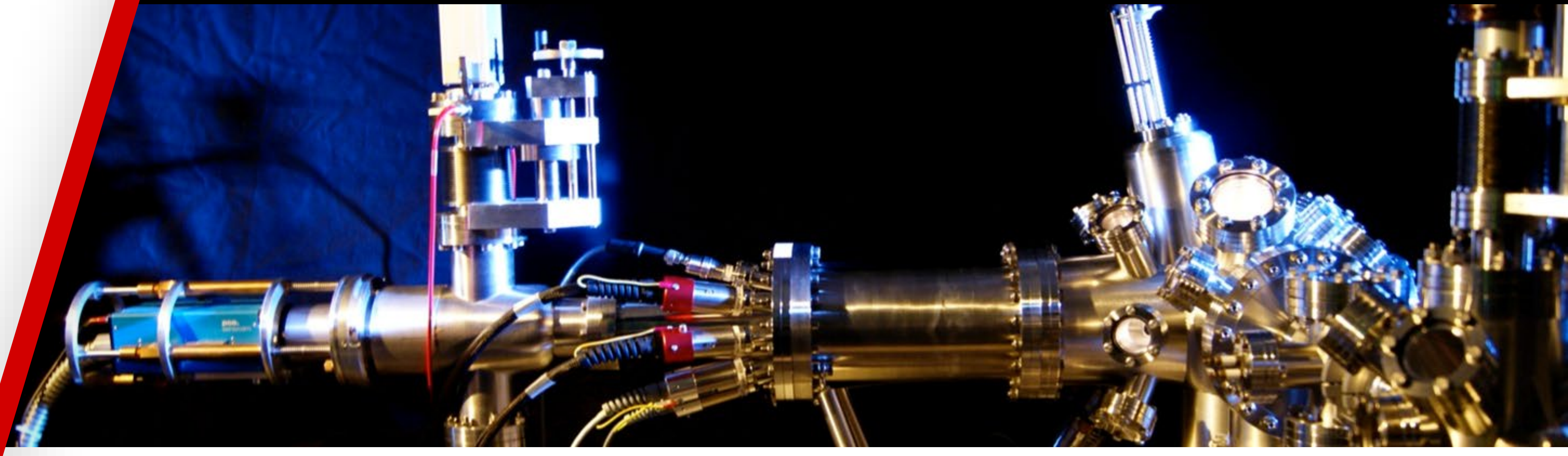
Mixing the Light- Spin with Plasmon-Orbit by Non-Linear Light Matter Interaction

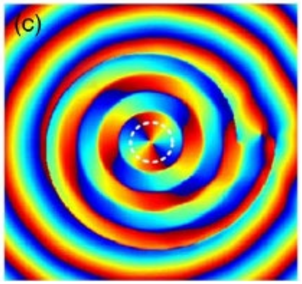


M. Aeschlimann
University of Kaiserslautern-Landau, Germany

Outline

Introduction

- What is the orbital angular momentum (OAM) of light?
- How can we create plasmon (SPP) with OAM?
- Time-resolved interferometric PEEM technique

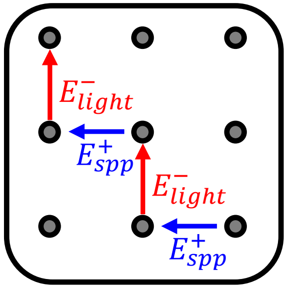


Dynamics of OAM in plasmonic vortices

- Real time view of a spiral phase
- Time domain simulation

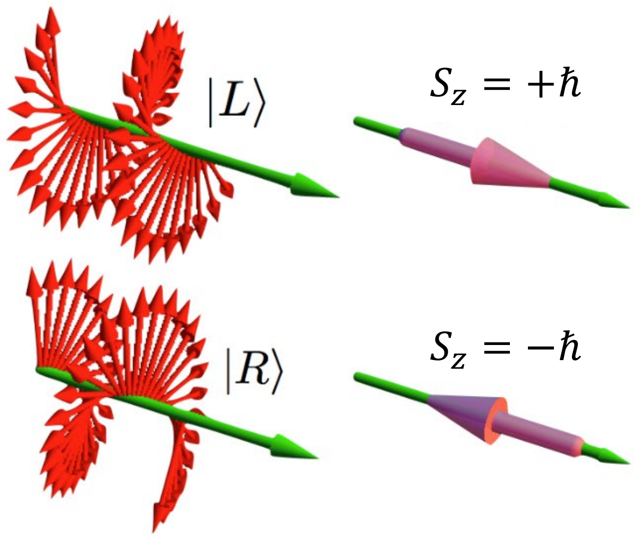
Demonstration of nonlinear optical spin-orbit conversion

- What do we really observe using nonlinear PEEM



Each photon has a spin

Photons are bosons



$$|L\rangle = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ i \\ 0 \end{pmatrix} e^{i(kz-\omega t)}$$

$$|R\rangle = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ -i \\ 0 \end{pmatrix} e^{i(kz-\omega t)}$$

Can we get a higher angular momentum?

Yes, with orbital angular momentum (OAM)!

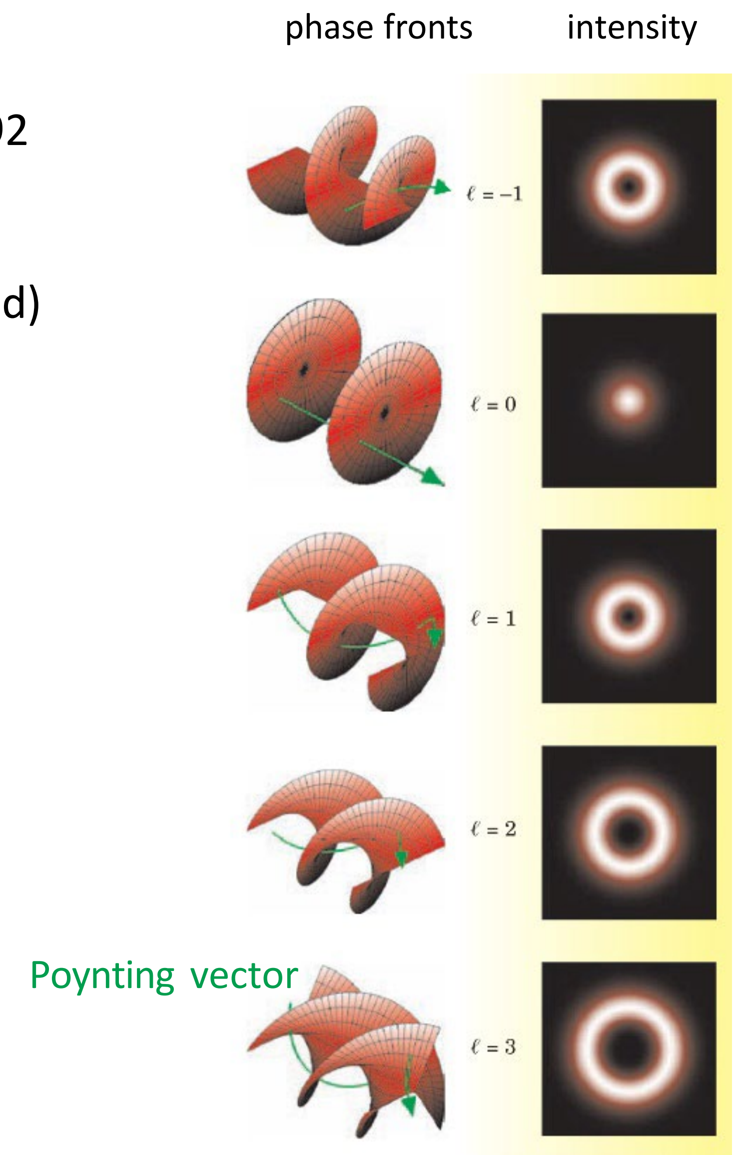
https://en.wikipedia.org/wiki/Spin_angular_momentum_of_light

Orbital angular momentum (OAM) of light

- Theoretical prediction by Allen et al. in 1992
- OAM: $L_z = l\hbar$, $\boldsymbol{l} \in \mathbb{Z}$ (l = 10‘000 realized)
- Helical wavefronts

  $\rightarrow \boldsymbol{E} = \boldsymbol{E_0} \cdot \boldsymbol{e^{-il\varphi}}$

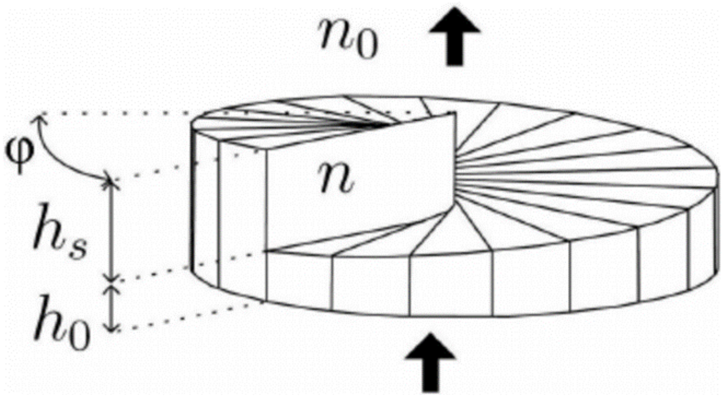
- Phase singularity on the optical axis
- OAM ≠ polarization!



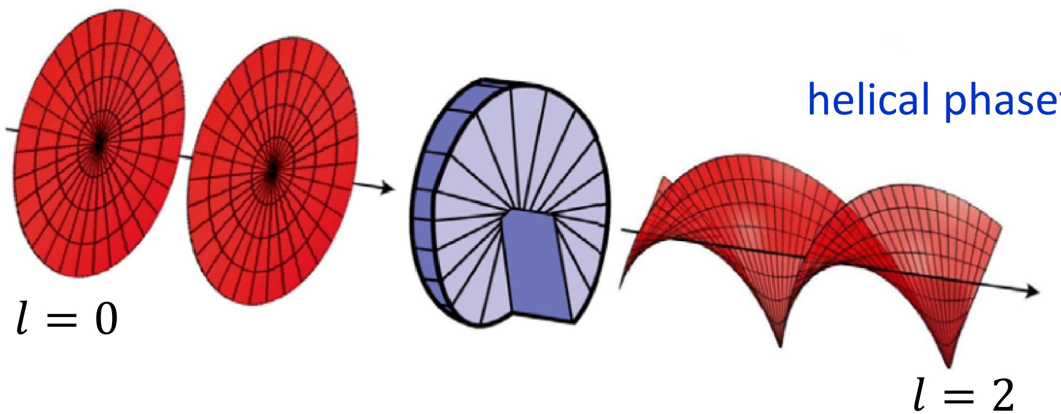
Padgett, Physics Today **57**, 35 (2004)
Yao & Padgett, Advances in Optics and Photonics **3**, 161 (2011)

Spiral phase plates

Thickness increasing linearly with the azimuthal angle



phase depends on the local thickness/optical path length



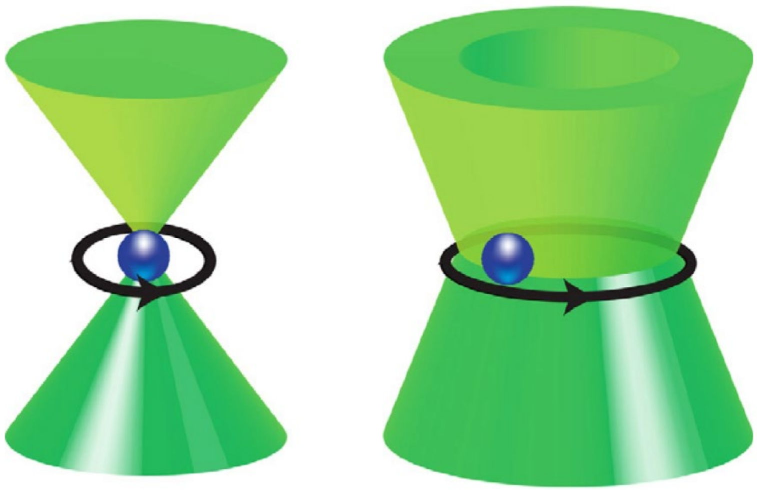
helical phasefront is created

Bovino, arXiv 1104.2201 (2011)
Yao & Padgett, Advances in Optics and Photonics **3**, 161 (2011)

Applications of twisted light: Micro-manipulation

Optical tweezers with OAM light (1995):

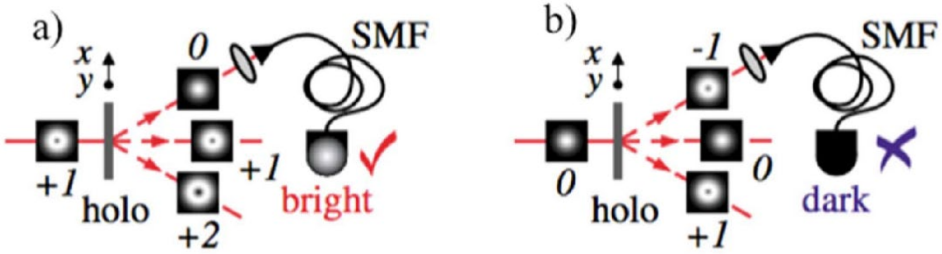
micro beads trapped by OAM light



He et al., Physical Review Letters **75**, 826 (1995)
Yao & Padgett, Advances in Optics and Photonics **3**, 161 (2011)
Jesacher et al., Optics Express **14**, 6342 (2006)

Applications of twisted light

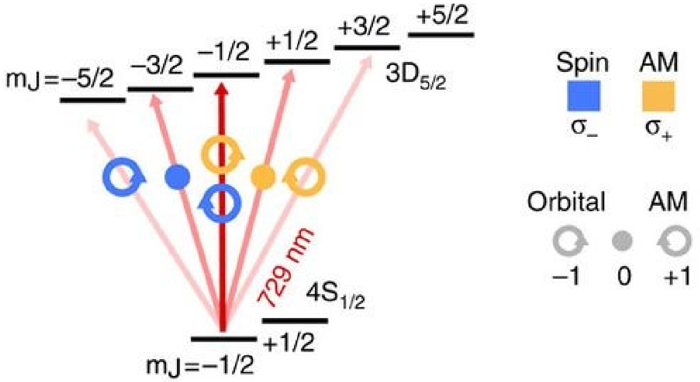
Quantum information processing



offers a new degree of freedom that can be used to encode information

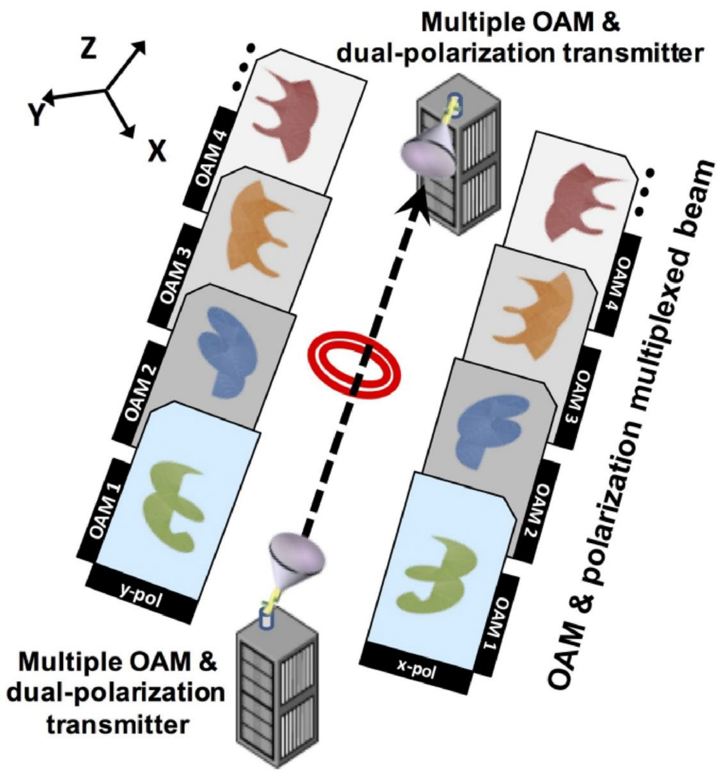
Erhardt et al., Light: Science & Applications **7**, 17416 (2018)

Fundamental research with single atoms



Rubinzstein-Dunlop et al., J. Opt. **19** (2017)
Schmiegelow et al., Nat. Comm. **7** (2016)

Optical communication

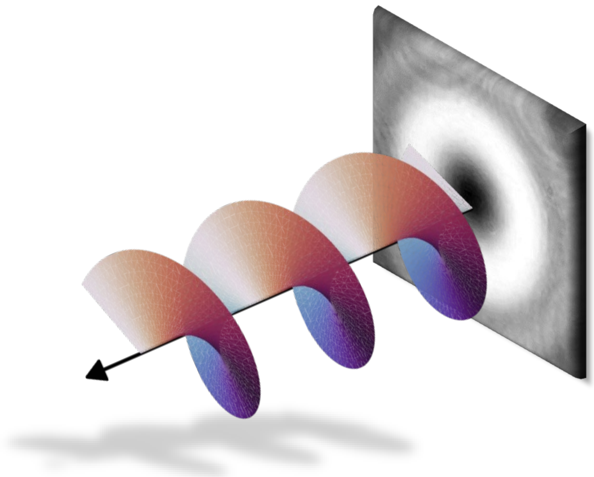


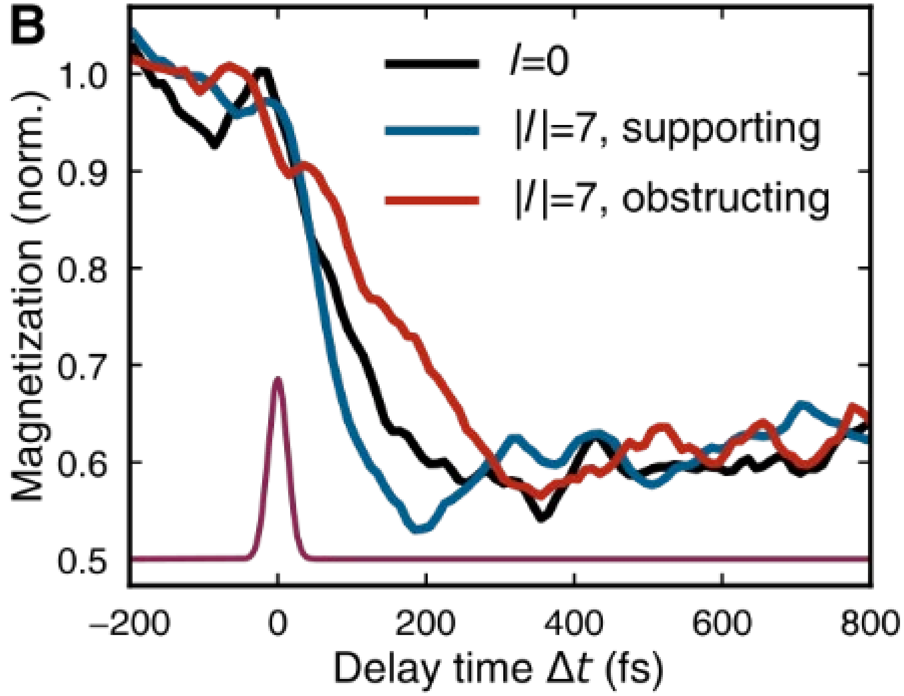
Wang, Photonics Research **4**, B14-B28 (2016)
Rubinzstein-Dunlop et al., J. Optics **19** (2017)

Solid state applications of twisted light ?

manipulation of magnetism with OAM light

Prinz et al, arXiv:2206.070502



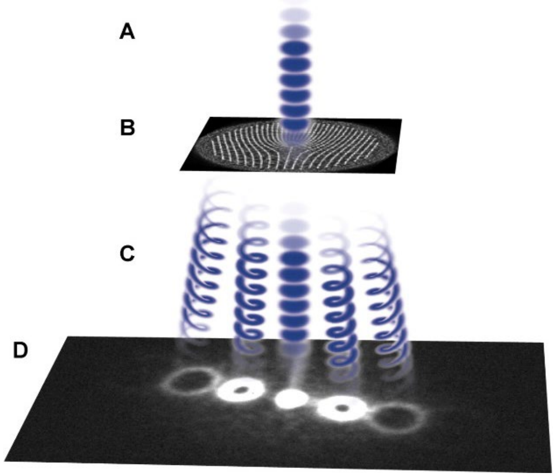


twisted light affects ultrafast demagnetization

but a direct transfer of orbital angular momentum can be ruled out

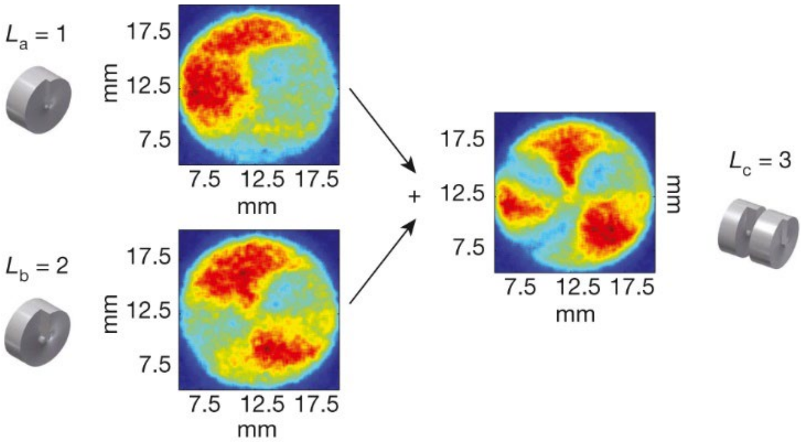
OAM beyond electro-magnetic waves

Electrons:



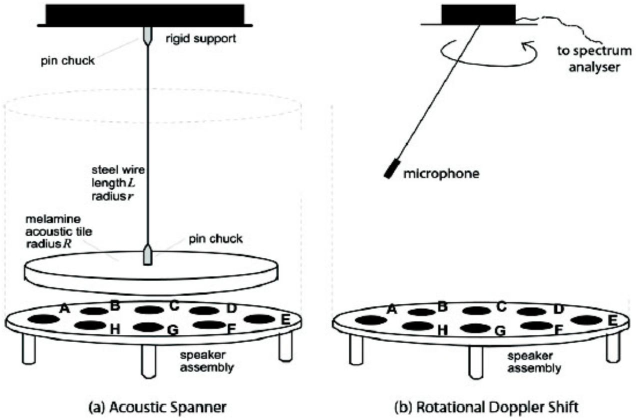
McMorran et al., Science **331**, 192-195 (2011)

Neutrons:



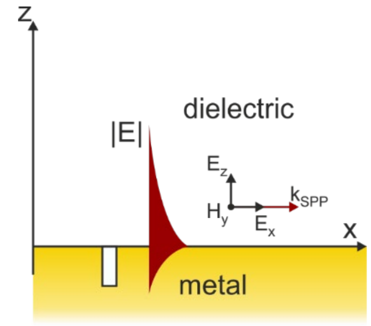
Clark et al., Nature **525**, 504-506 (2015)

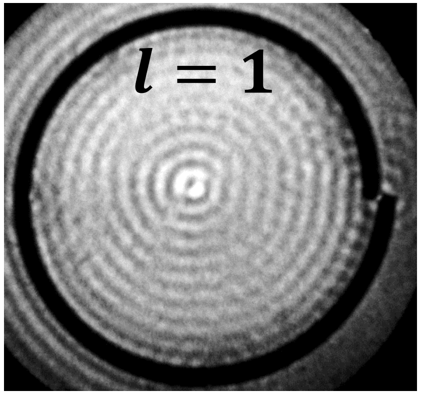
Sound waves:



Skeldon et al., NJP **10**, 013018 (2008)

Surface plasmon polaritons (SPPs):





Gorodetski et al., PRL **101**, 043903 (2008)
Spektor et al., Science **355**, 1187 (2017)

Collaboration

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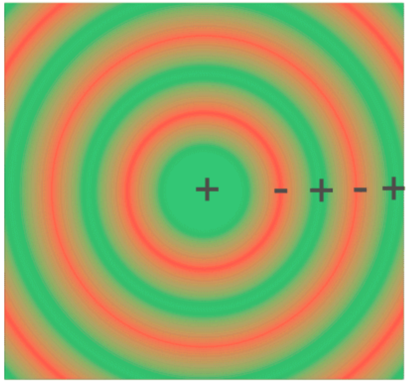
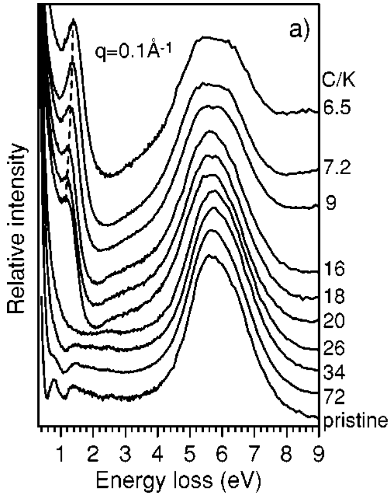
Eva Prinz
Michael Hartelt
Tobias Eul
Deirdre Kilbane

University of Kaiserslautern, Germany

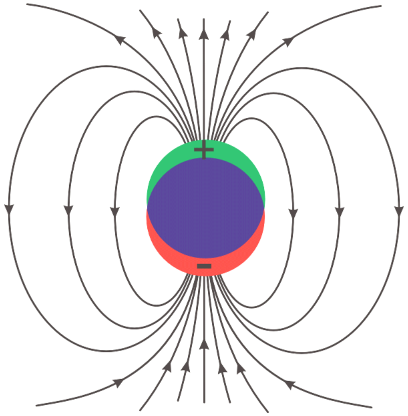
Plasmonic modes

Volume plasmon

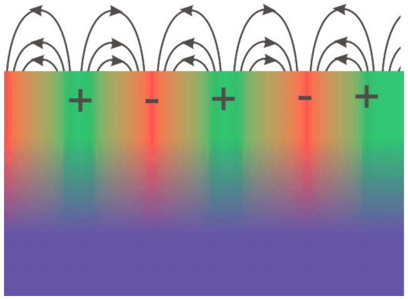


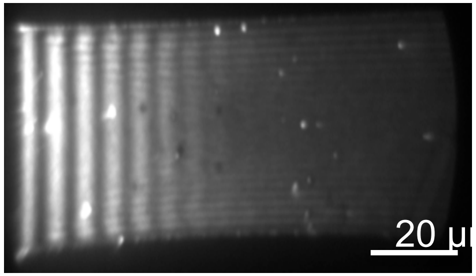


Particle plasmon



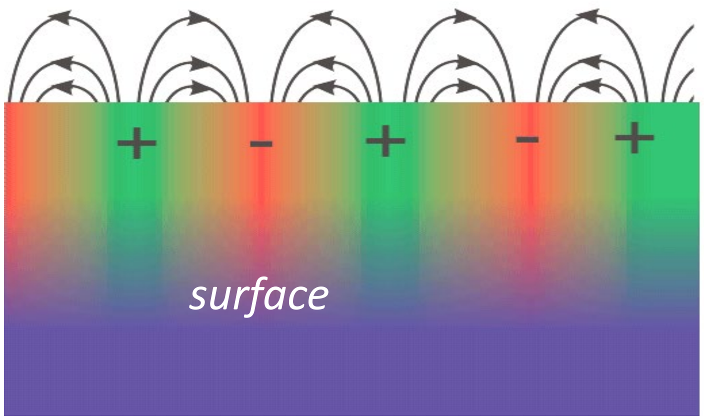
Surface plasmon polariton



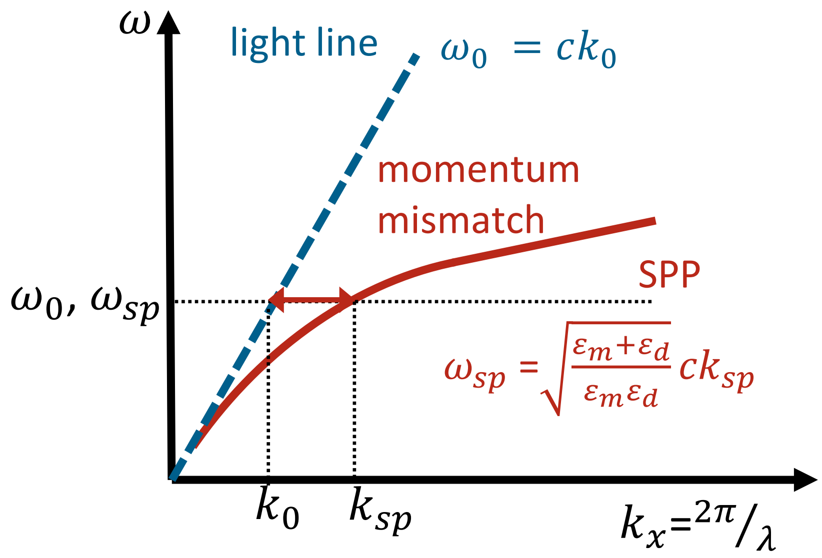


Surface plasmons polaritons (SPP)

hybrid modes of a **light field** coupled to a **coherent longitudinal electron oscillation** propagating along the interface of a metal and a dielectric



Dispersion relation



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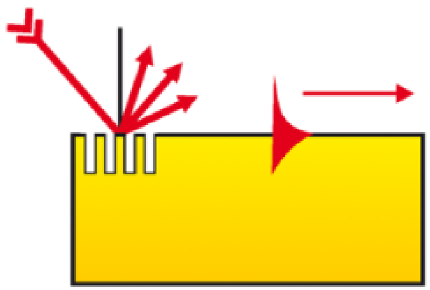
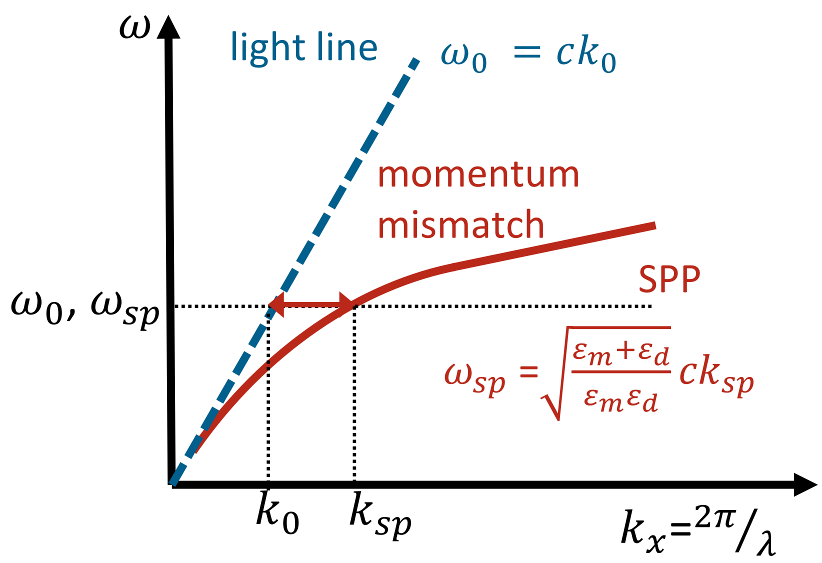


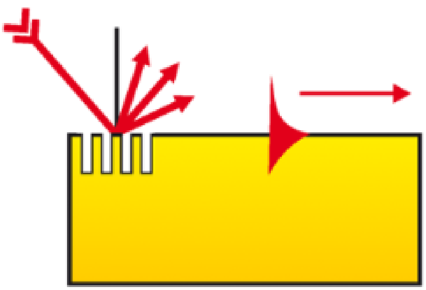
- Optical grating or simple edge provides the missing momentum

Dispersion relation



Surface plasmons polaritons (SPP)

hybrid modes of a **light field** coupled to a **coherent longitudinal electron oscillation** propagating along the interface of a metal and a dielectric



- Optical grid or simple edge provides the missing momentum

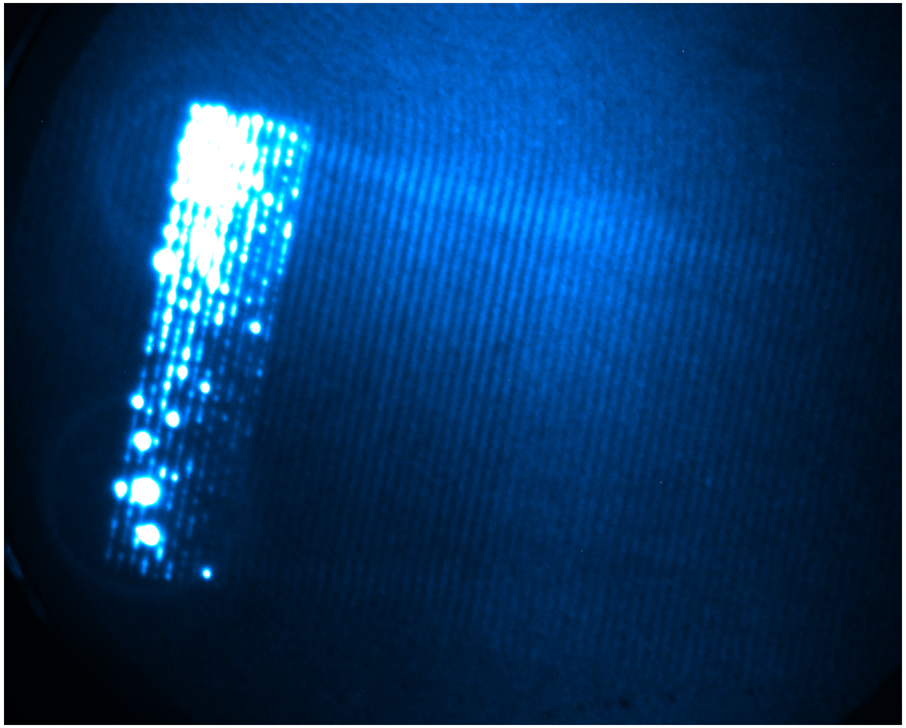
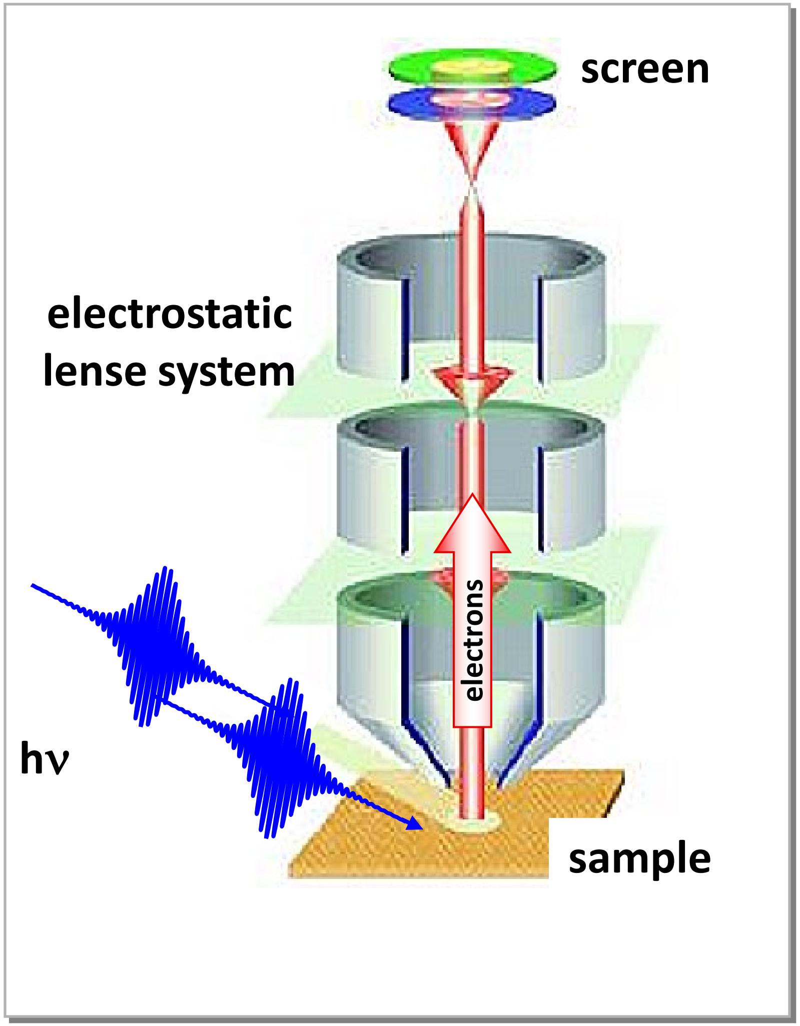
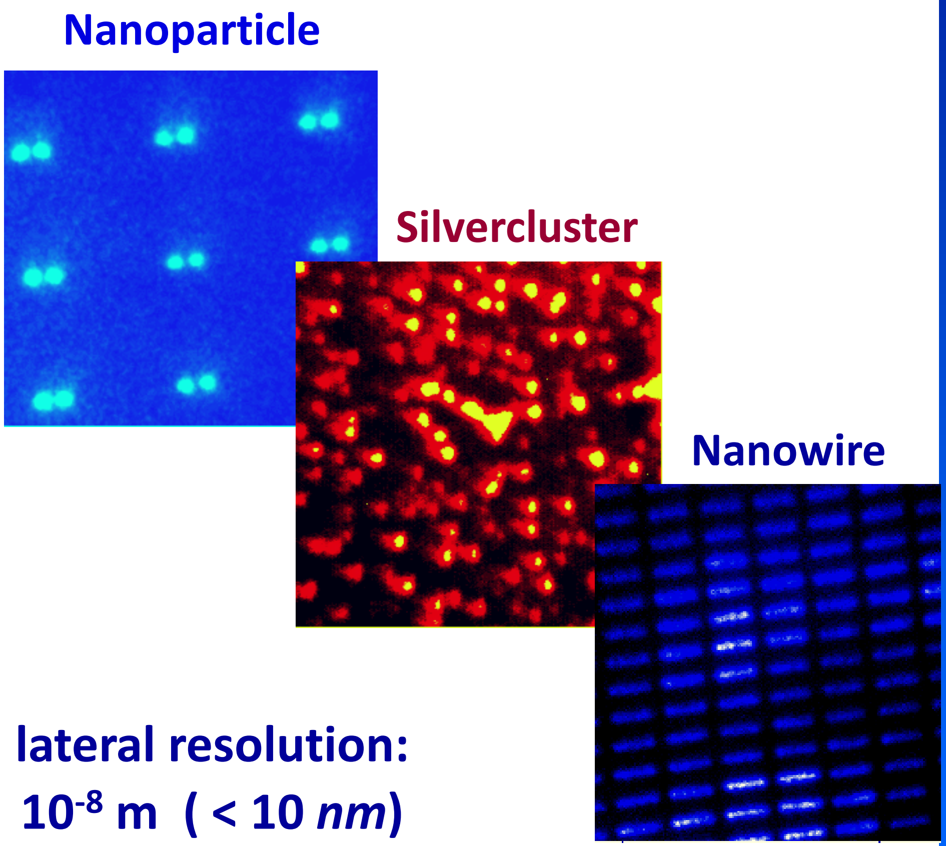


Photo Emission Electron Microscopy: PEEM

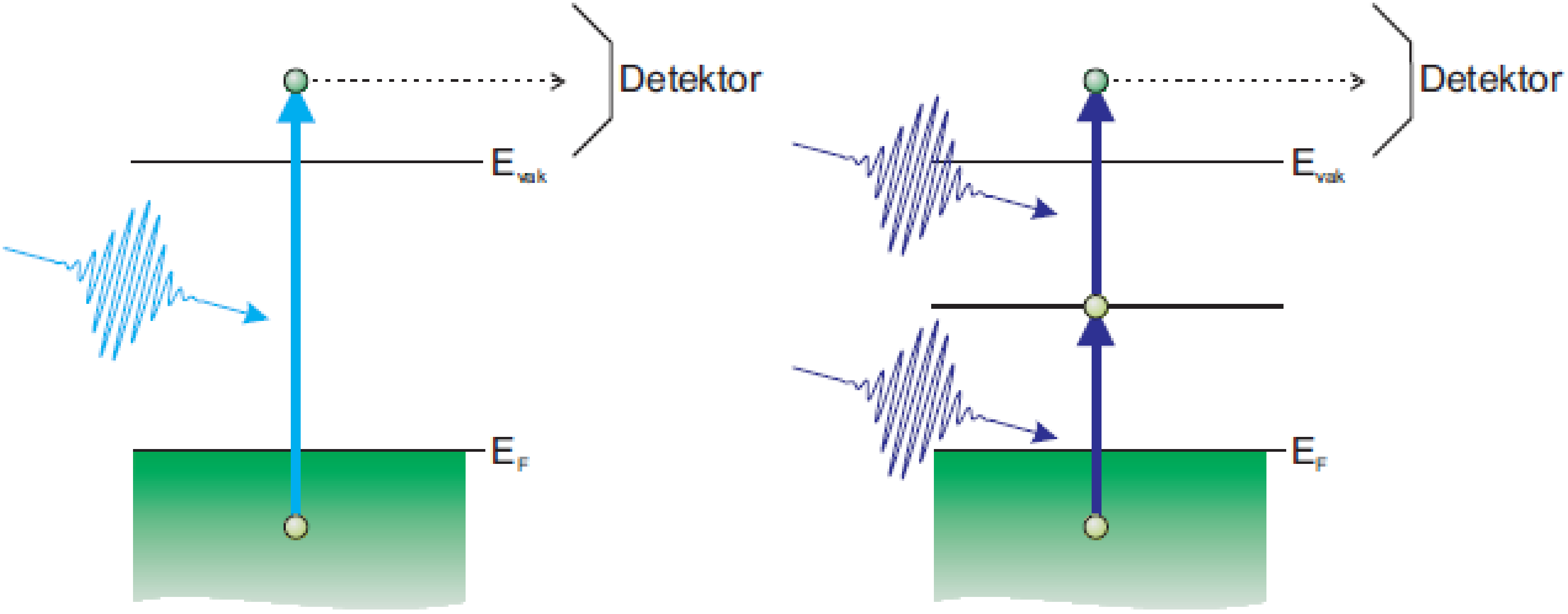




lateral resolution:
10^{-8} m (< 10 *nm*)

time resolution:
< 10^{-15} sec (< 1 *fs*)

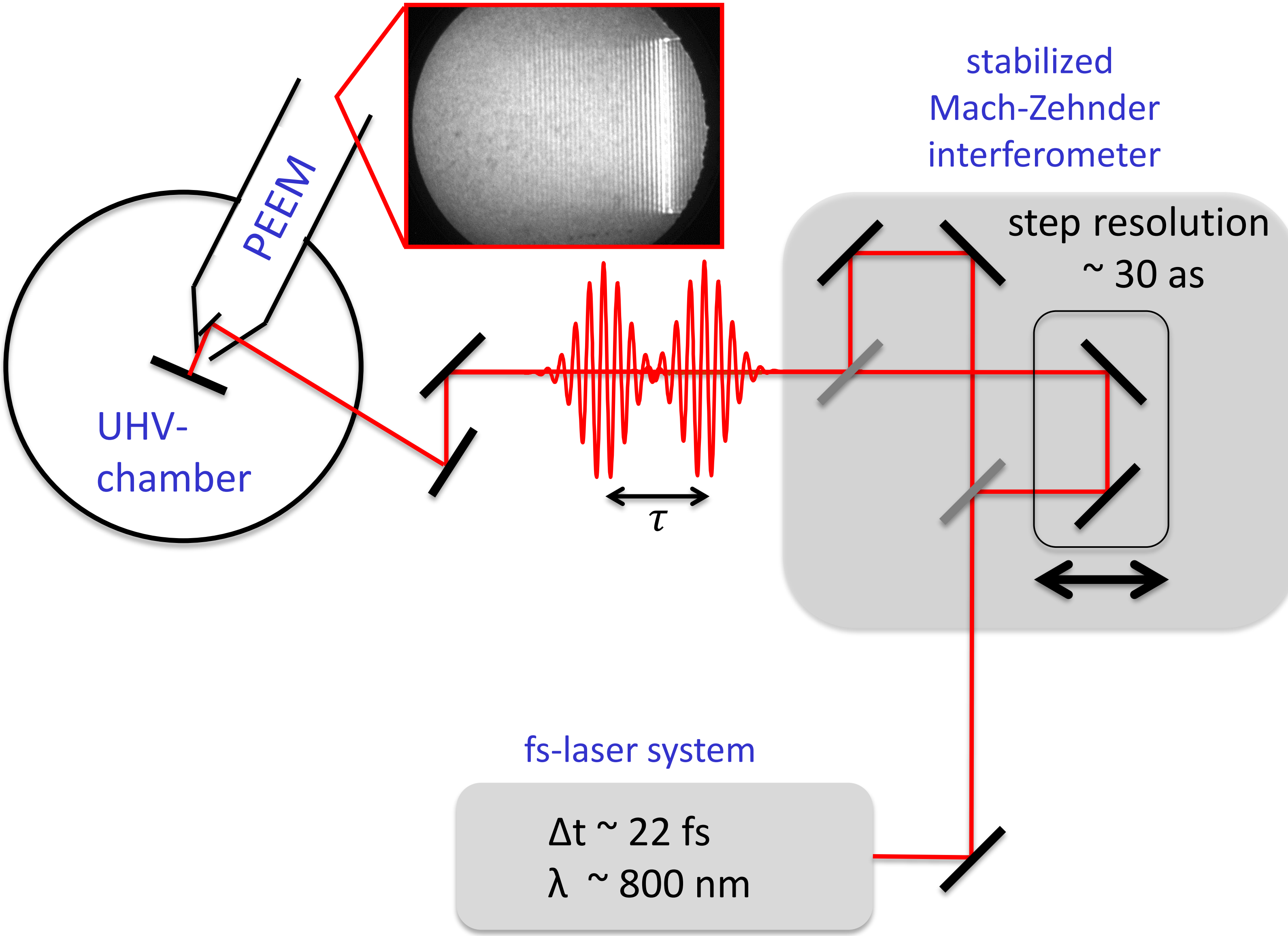
Plasmon driven electron emission



plasmon energies < 3 eV

typical workfunctions: 4eV - 6 eV

Interferometric time-resolved PEEM technique



Wehner et al., Opt. Lett. **22** (1997)

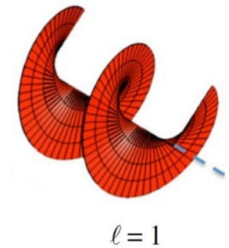
Light with a twist phase dislocation on an optical vortex

Spin (SAM)

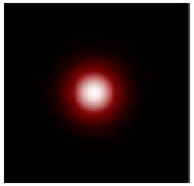
$S_z = \pm\hbar$

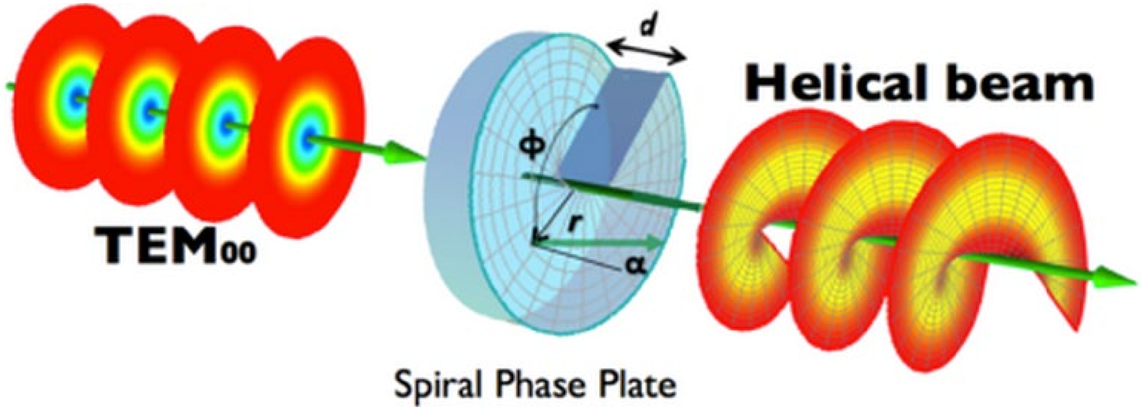
Light can carry angular momentum

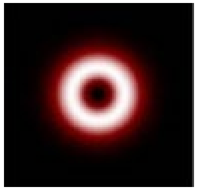
Orbital (OAM)



$L_z = \pm l\hbar$

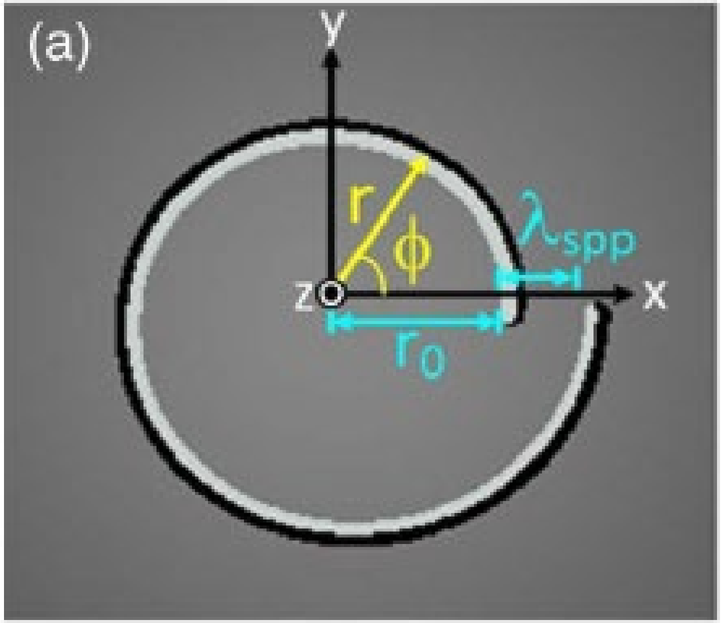




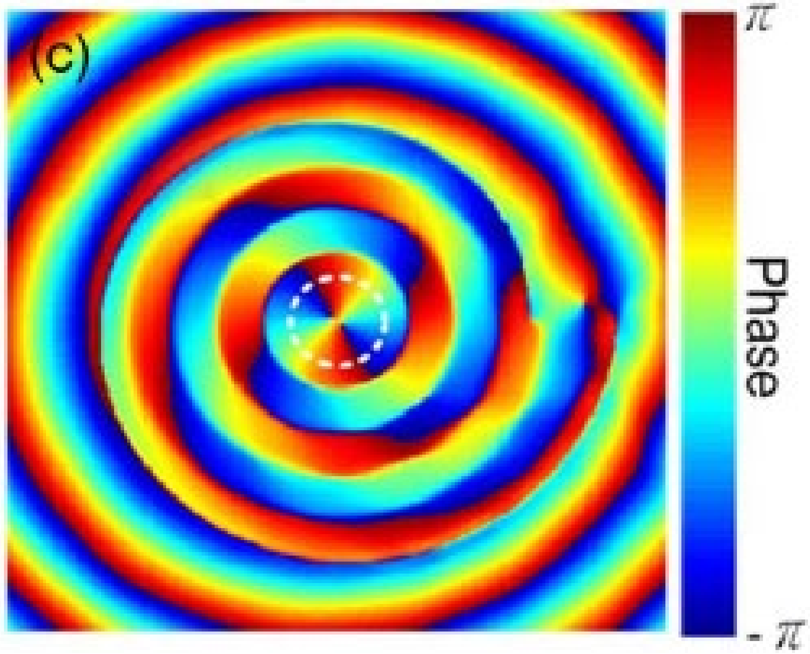


Can we do this with plasmonic waves?

Plasmonic Vortex Lens: Plasmonic Archimedes Spiral



spiral phase profile

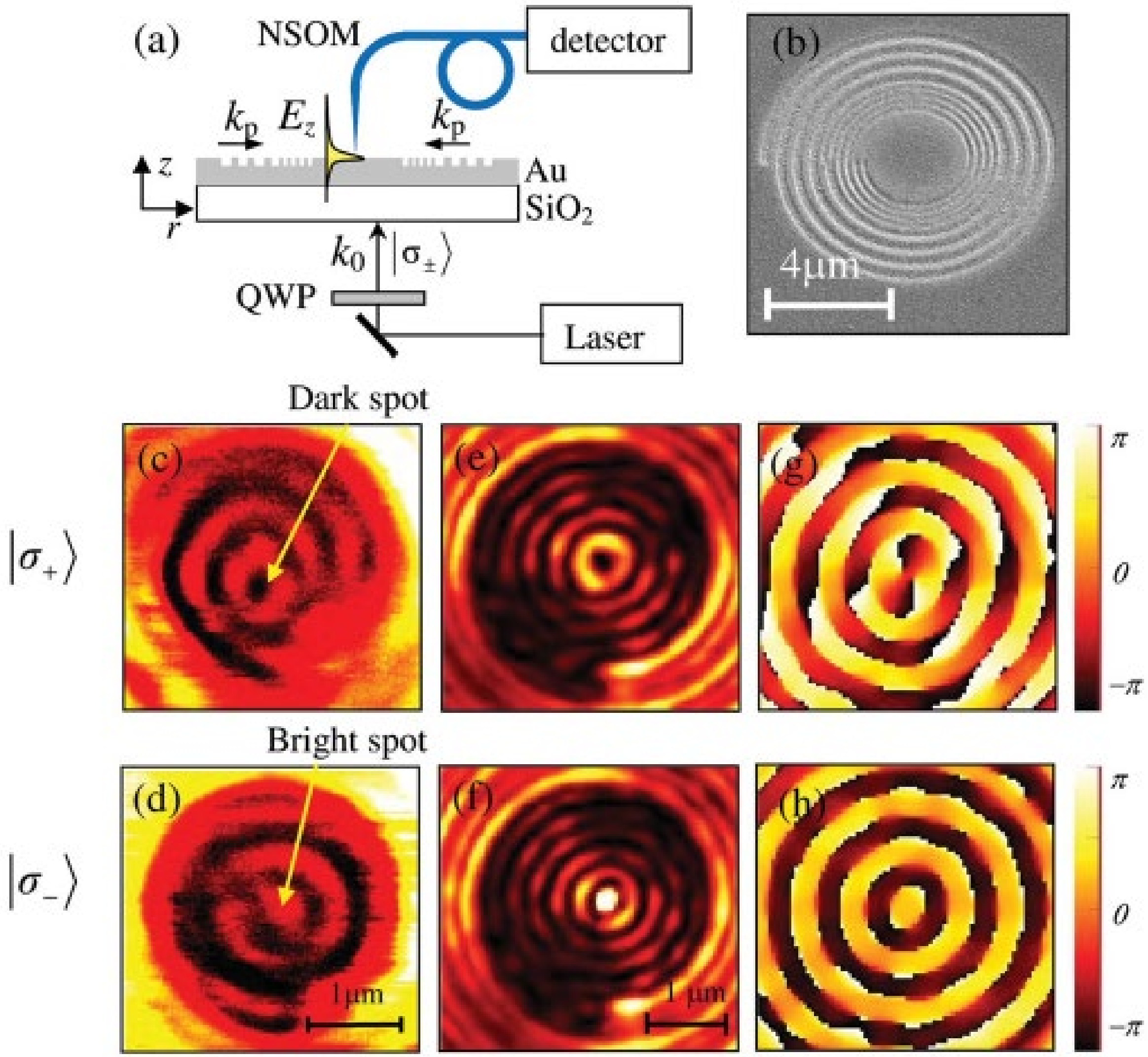


Phase singularity & rotational flow

$$r(\phi) = r_0 + \frac{(m\phi\lambda_{spp})}{2\pi}$$

m = geometrical charge of the vortex

Plasmonic Vortex Lens: Pioneering work



dark spot

bright spot

Y. Gorodetski, A. Niv, V. Kleiner, and E. Hasman
Phys. Rev. Lett. **101**, 043903 (2008)

Topological Charge q: Optical Spin Orbit Coupling

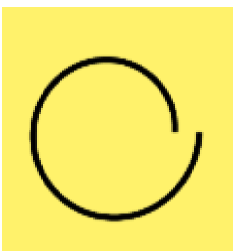
E. Hasman et al.
PRL **101**, 043903 (2008)

q = m+s

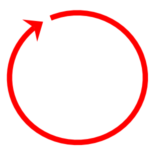
geometry spin

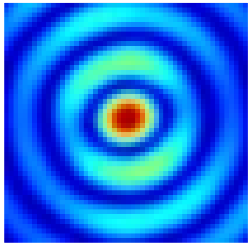
$$E \propto J_0\left(k_{spp} \cdot r\right)$$

m = 1



s = -1



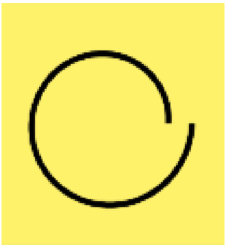


q = 0

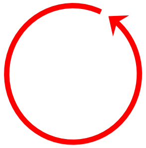
Focusing

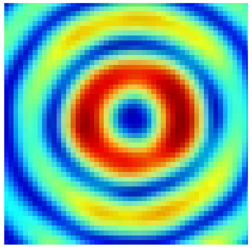
$$E \propto J_2\left(k_{spp} \cdot r\right)e^{j\mathbf{2}\theta}$$

m = 1



s = 1





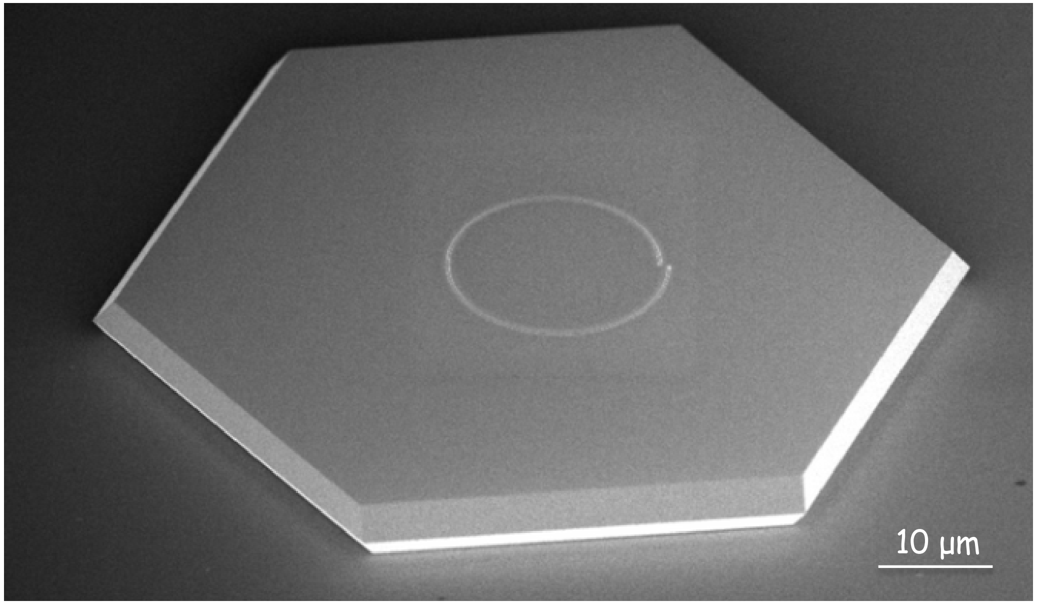
q = 2

Rotation

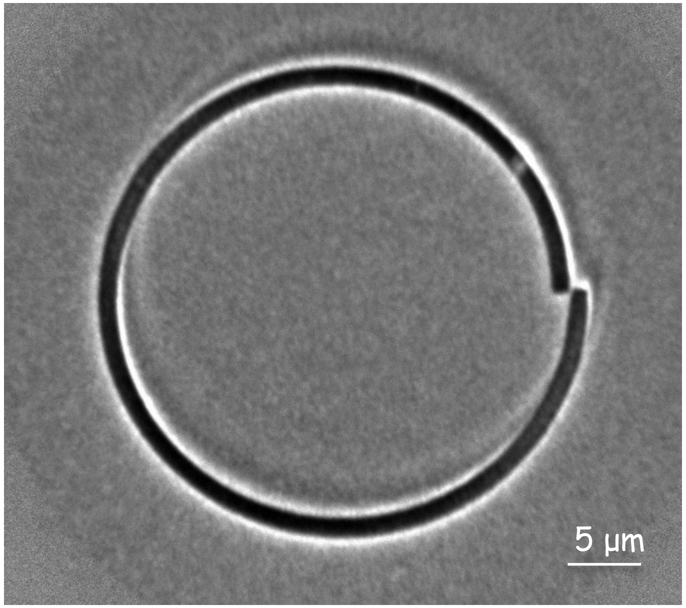
Plasmonic Vortex Lens preparation

- Depth 3 - 5 µm flakes grown on Si
- Focussed Ion Beam (FIB) milling of slits

SEM image



UV PEEM image

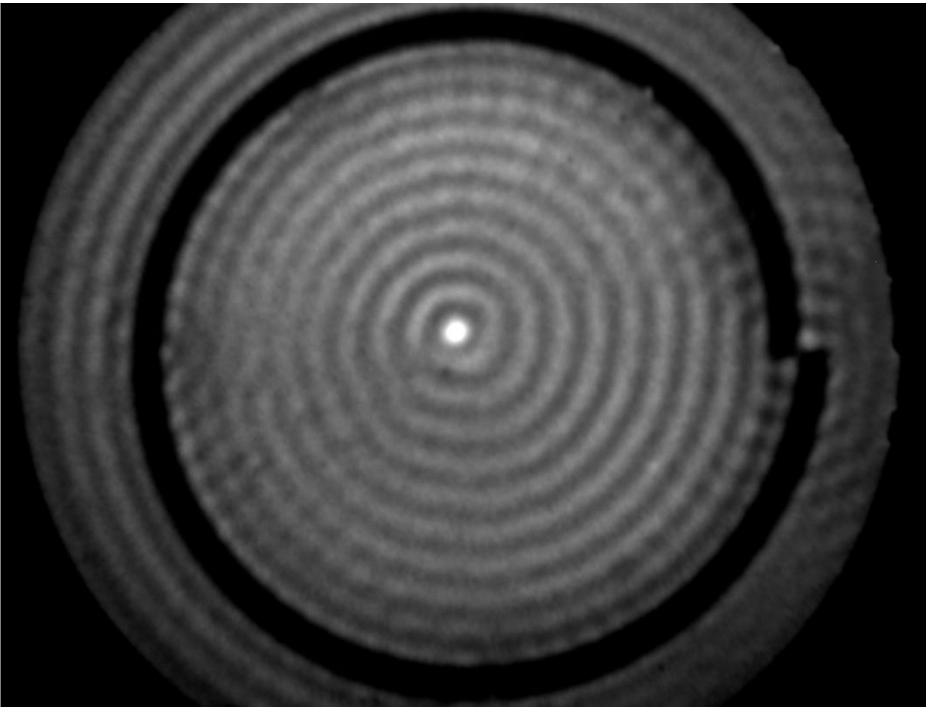


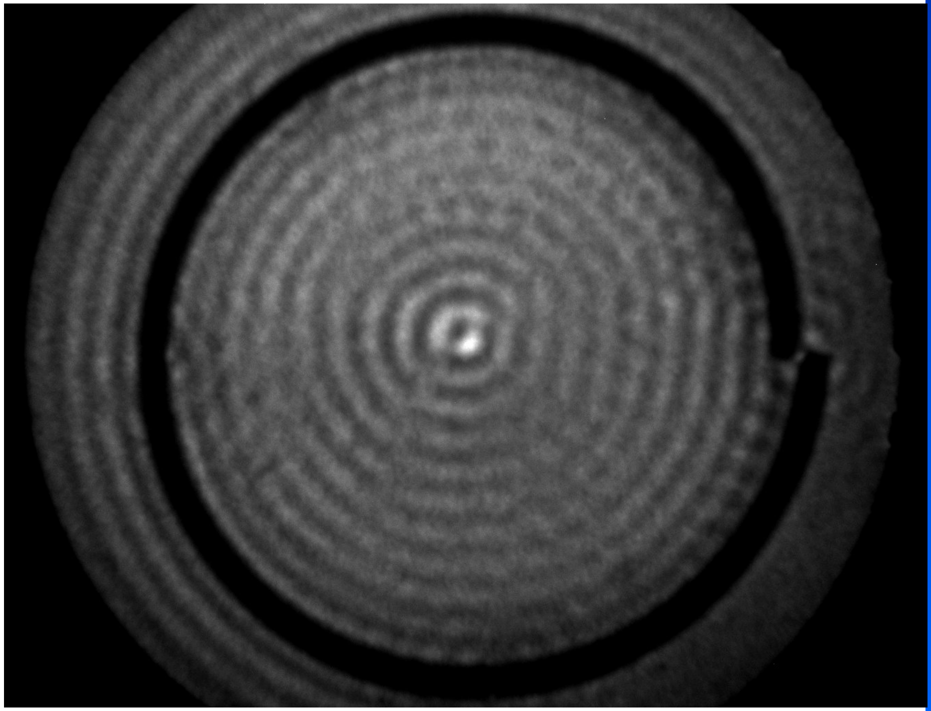
Spin-orbit coupling

q = m+s

s = -1

s = +1

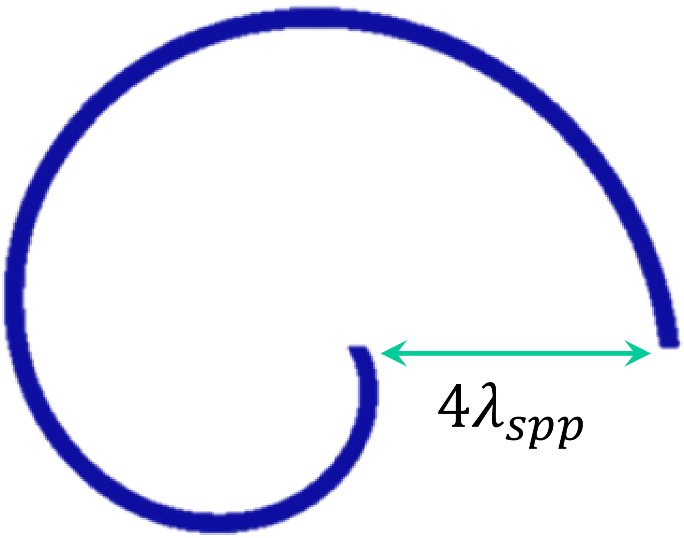




q = 1 - 1

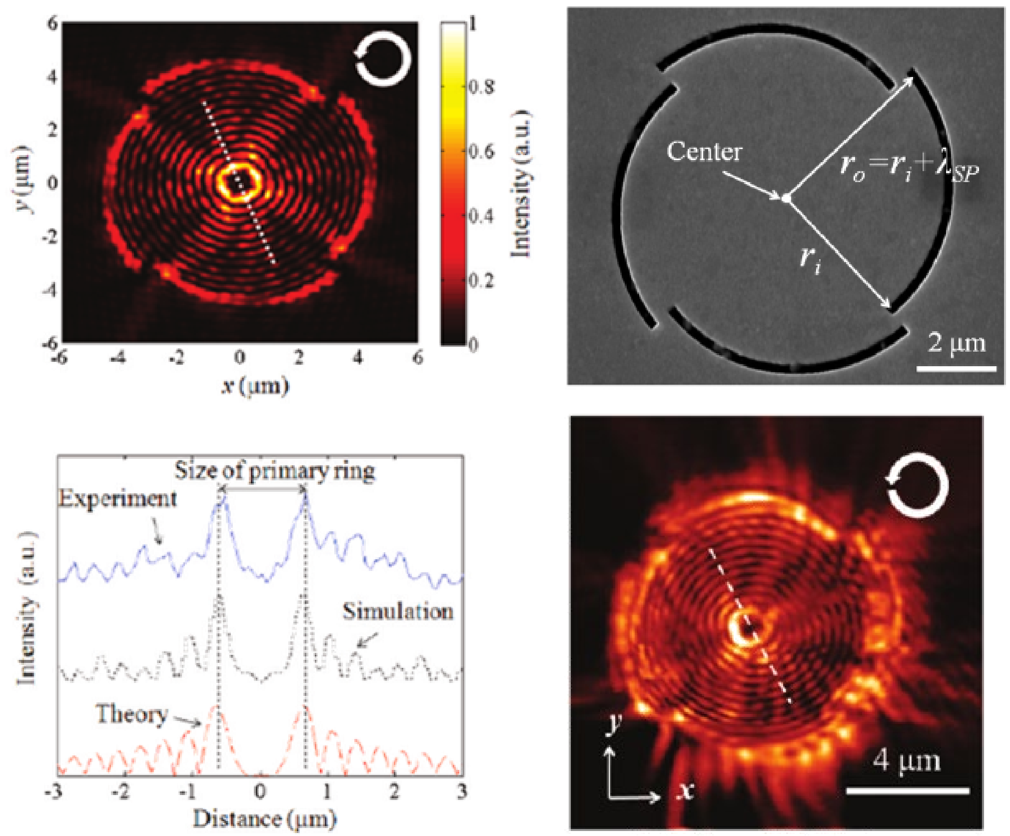
q = 1 + 1

High Order Plasmonic Vortex Lens



azimuthally
dependent losses

Segmented Spiral Structure

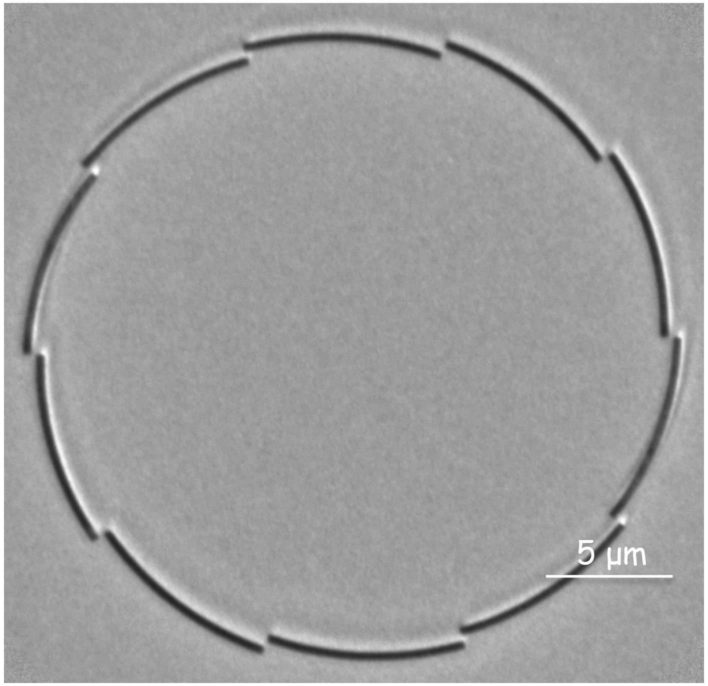


H. Kim, J. Park, S.-W. Cho, S.-Y. Lee, M. Kang, and B. Lee
Nano Letters 10, 529 (2010)

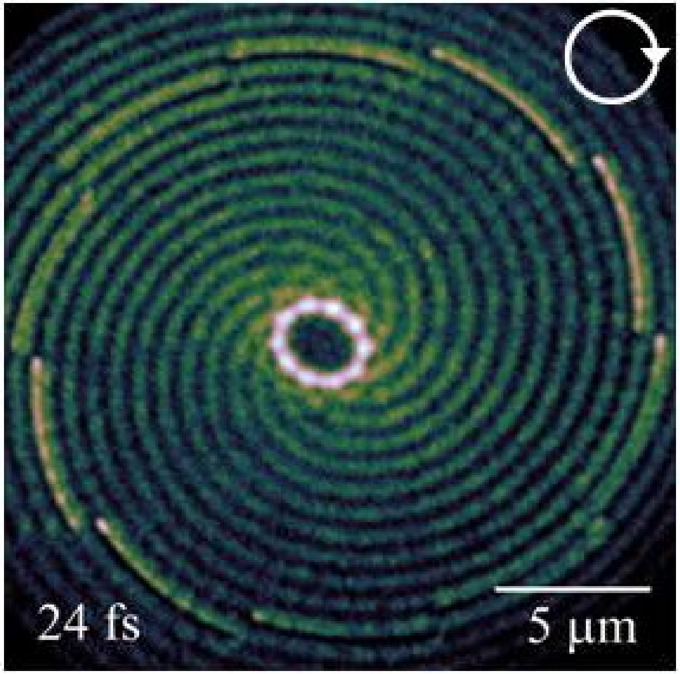
m = 10 plasmonic vortex lens

Atomically flat single crystalline Au flakes

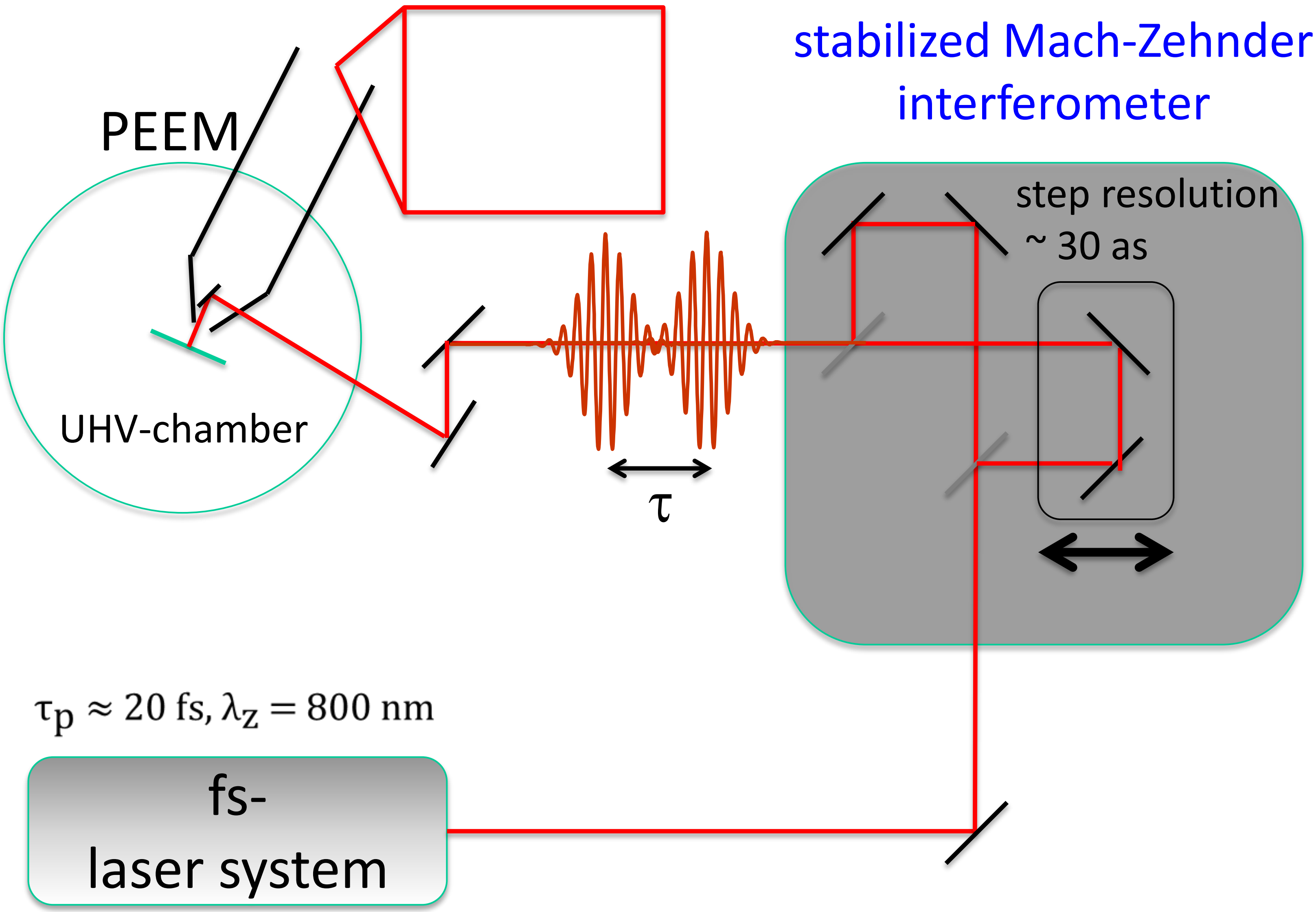
UV PEEM image



800nm PEEM image

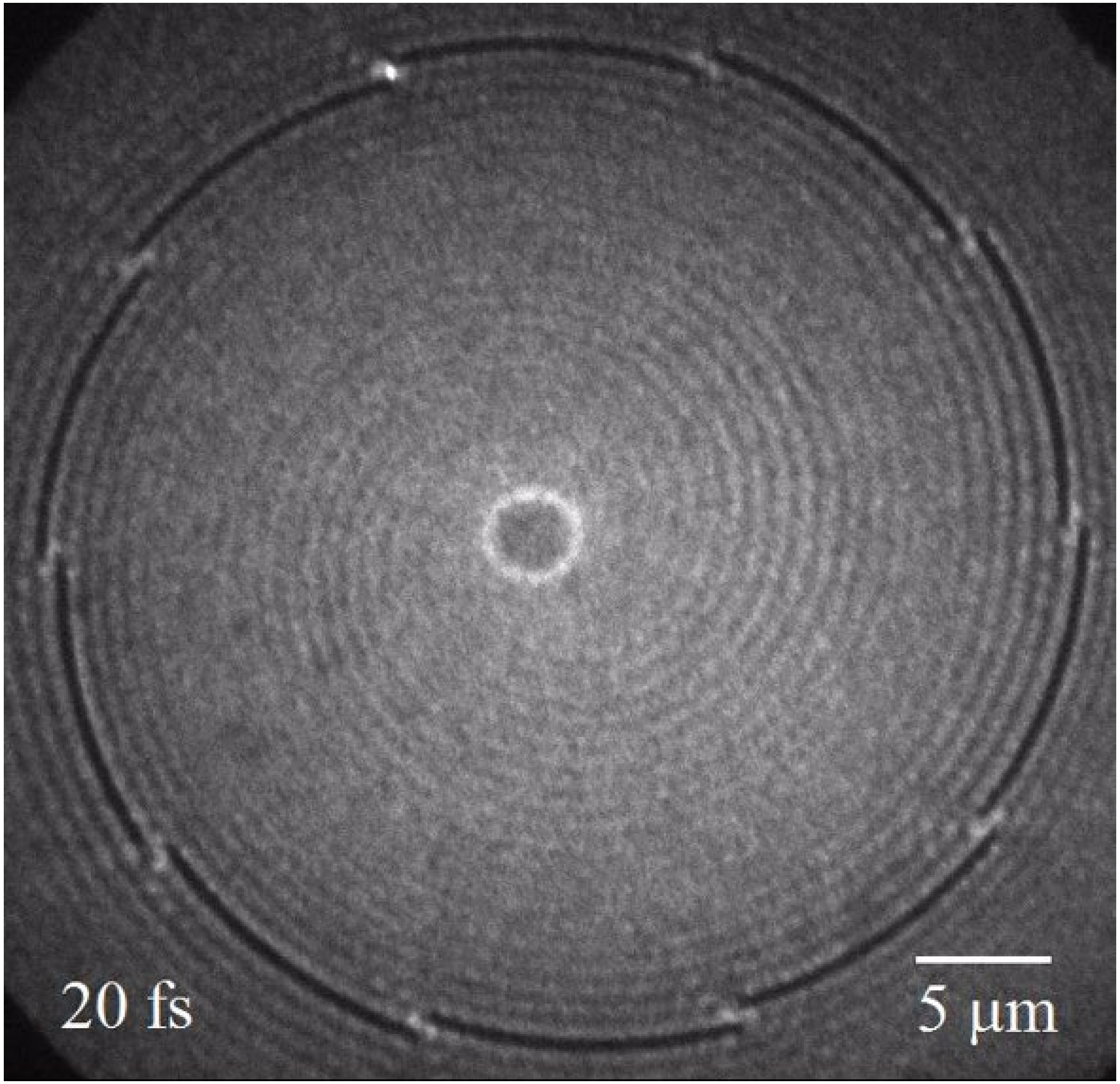


Interferometric time-resolved PEEM technique



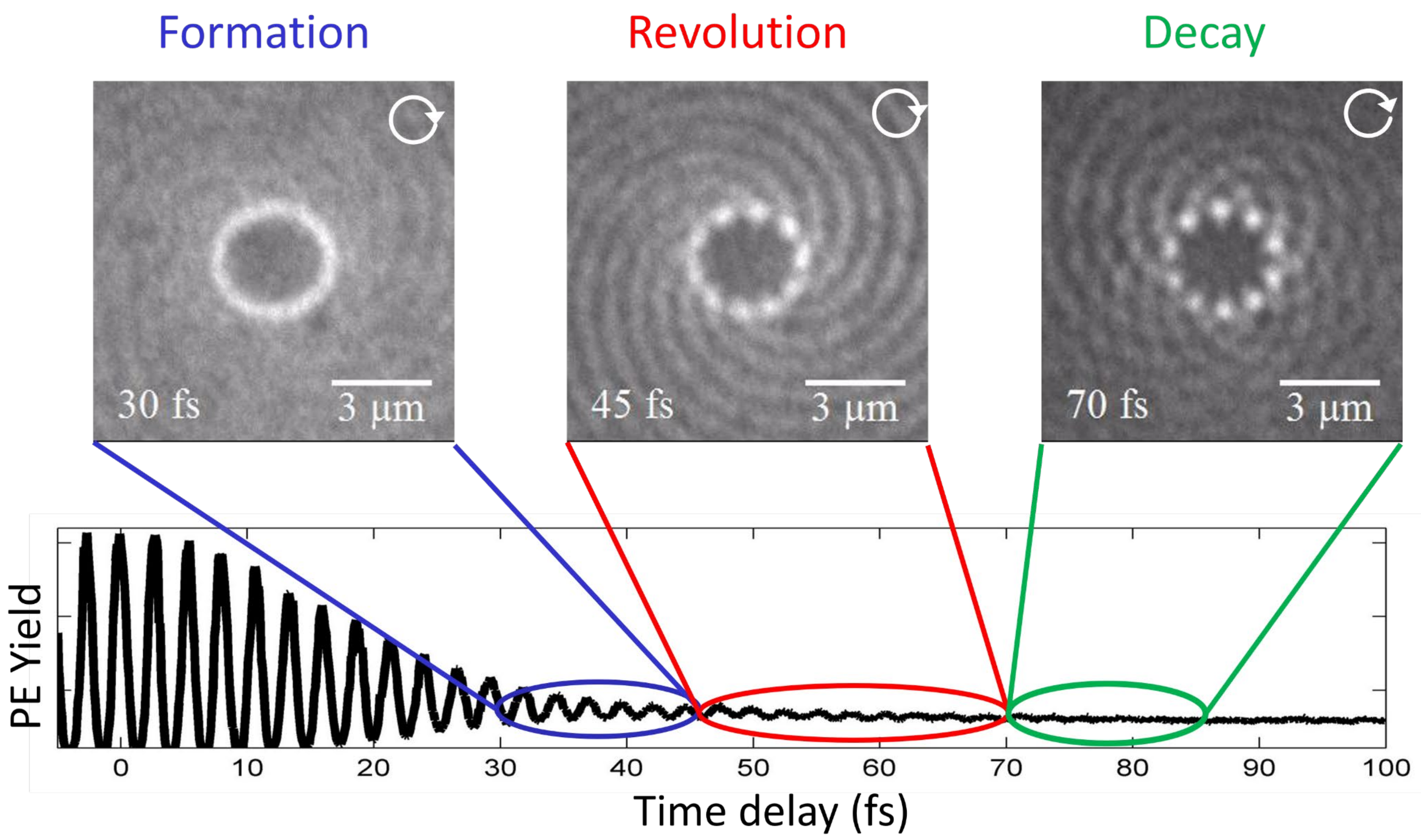
Appl.Phys.B **74** (2002) 223

TR-PEEM movie: ultrafast dynamics of a plasmonic vortex



Science 355, 1187 (2017)

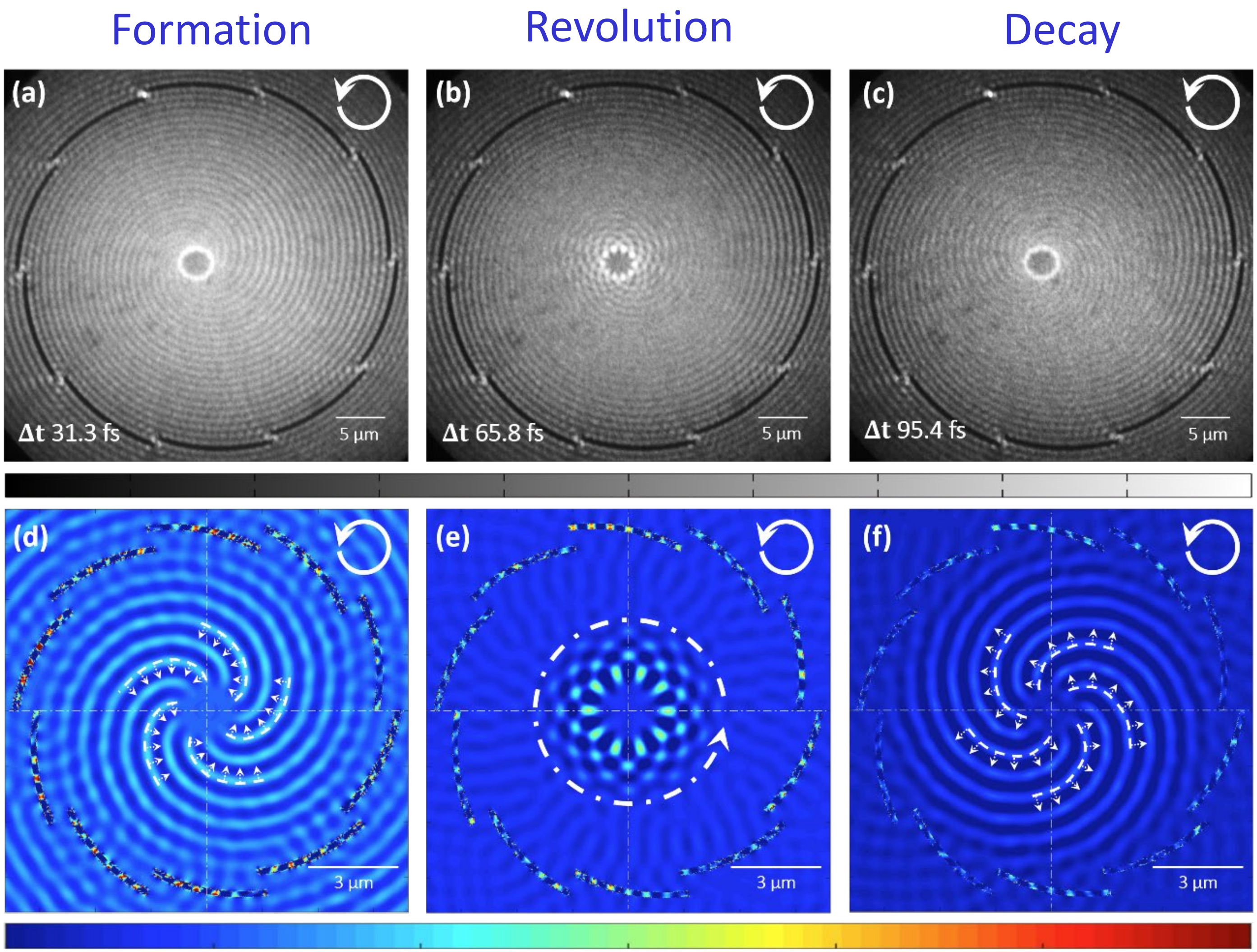
Lifetime of a plasmonic vortex



Experiment & Simulation

Grisha Spektor

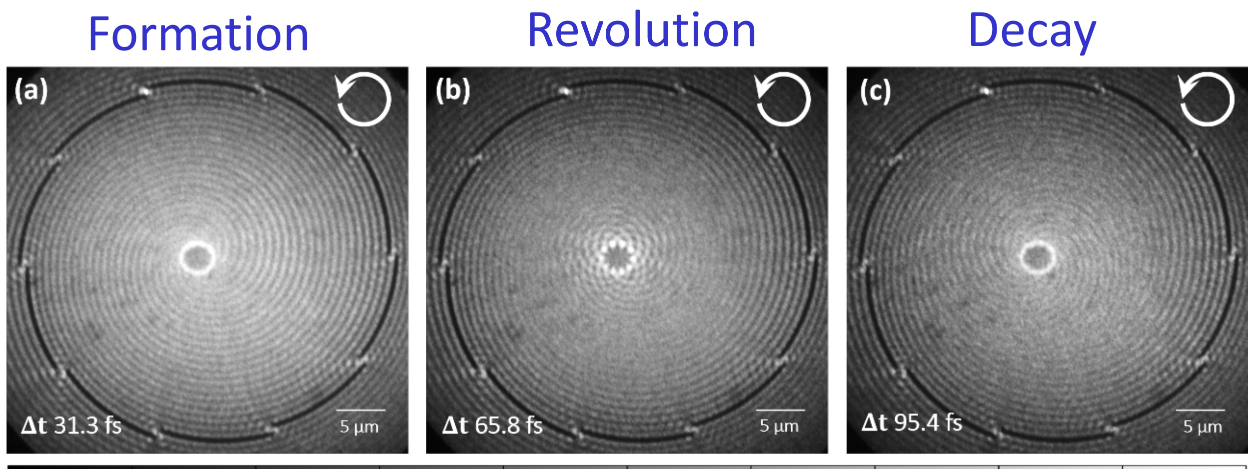
Experiment



FDTD Lumerical Simulation

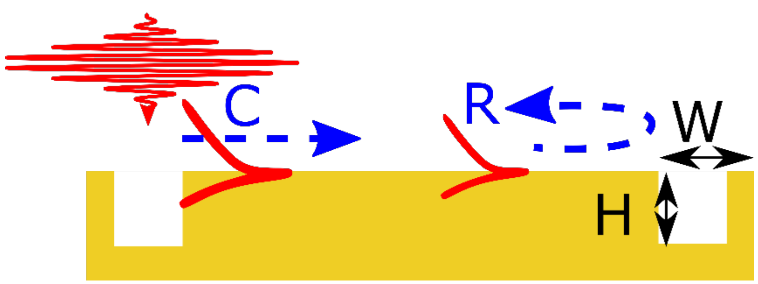
Science 355, 1187 (2017)

Reflection from boundaries



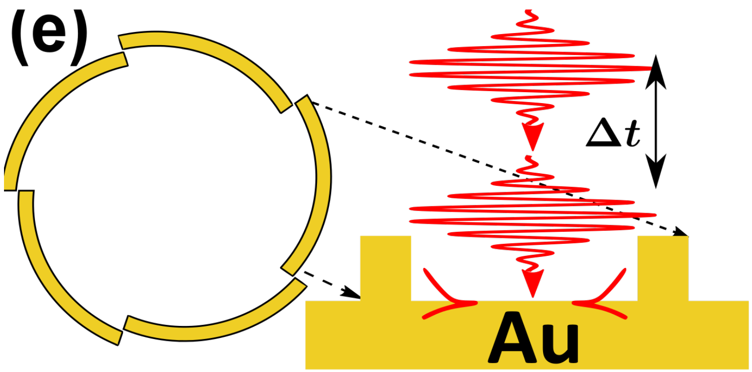
After n reflections:

$$l_{\mathrm{SPP}} = (m + s_{\mathrm{light}}) + 2 \cdot m \cdot n$$

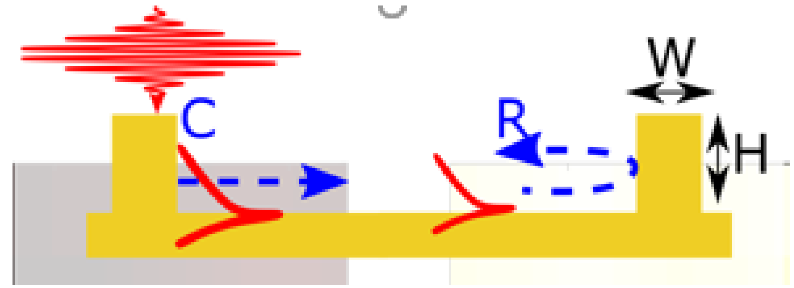


Spektor, Prinz et al., Science Advances **7**, 33 (2021)

Chiral cavity reflectors: ridges instead of slits



plasmonic chiral cavity of order *m=5*

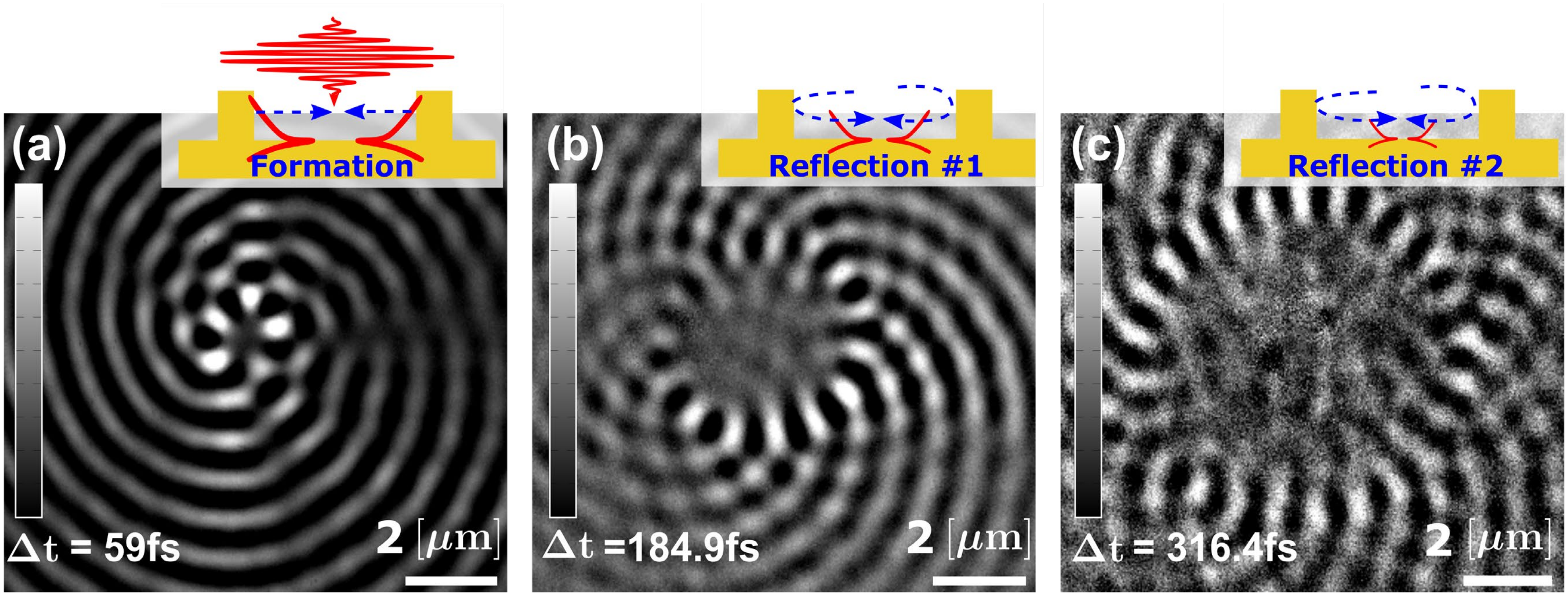


ridge design facilitates ~95% reflectivity

OAM of the order $l=m+2\cdot m$

Chiral cavity reflectors

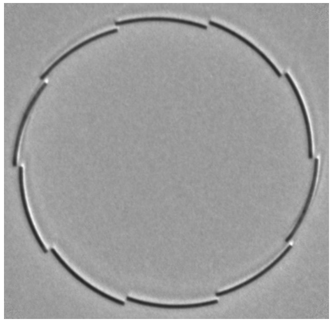
OAM of the order *l=m+2·m*



$l_0 = 5$ $l_1 = 15$ $l_0 = 25$

Sci. Adv. 2021, 7, eabg5571 (2021)

Lobe Angular Velocity

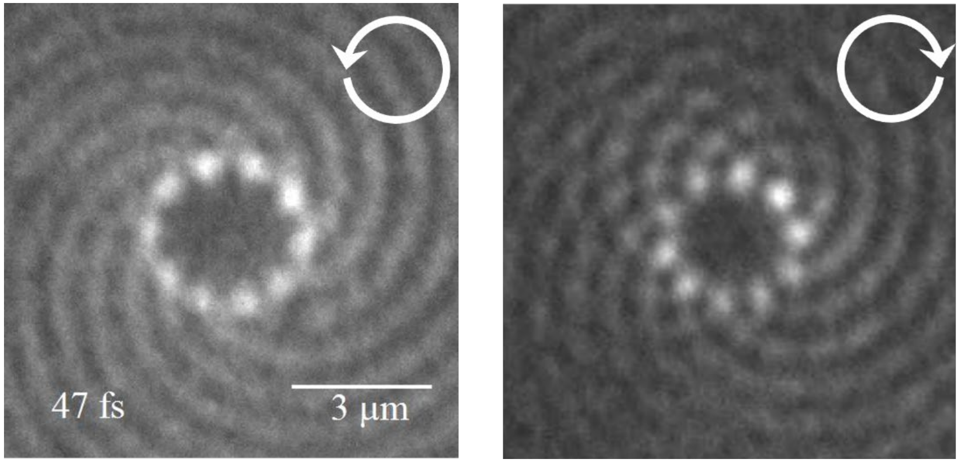


m = 10 plasmonic vortex lens

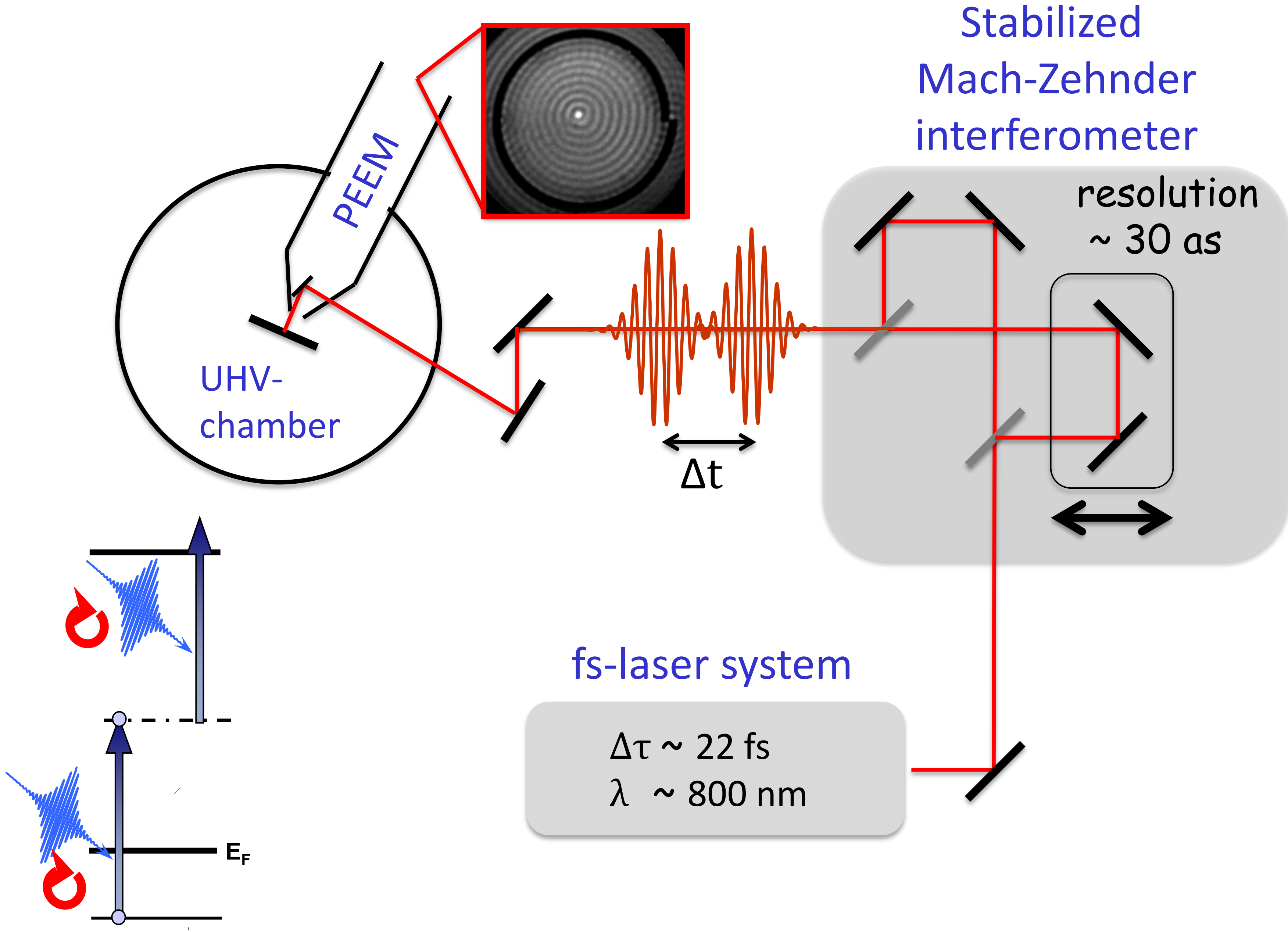
$$l = m + s$$

$$l = 11 \qquad l = 9$$

- **# of lobes**?
- Radial dependence?
- Angular dependence?



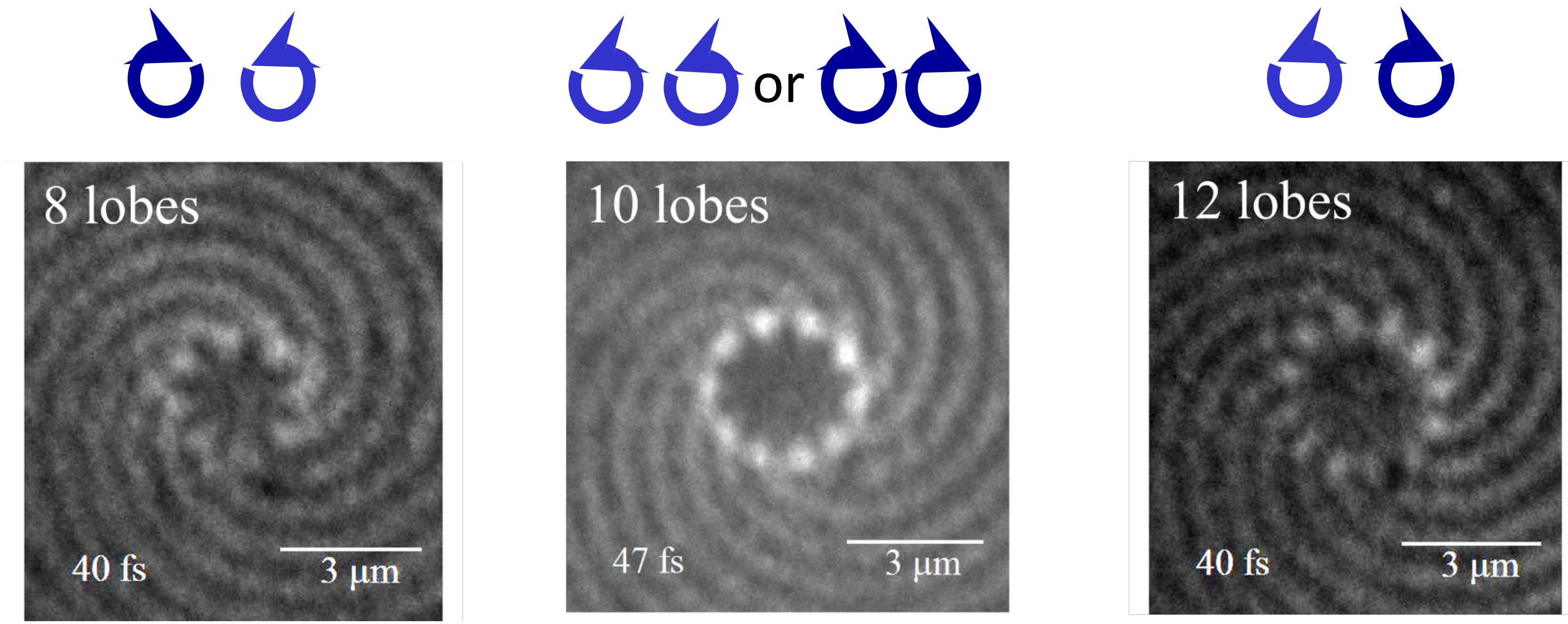
Time-resolved NI-PEEM



Subtractive spin-orbit mixing process?

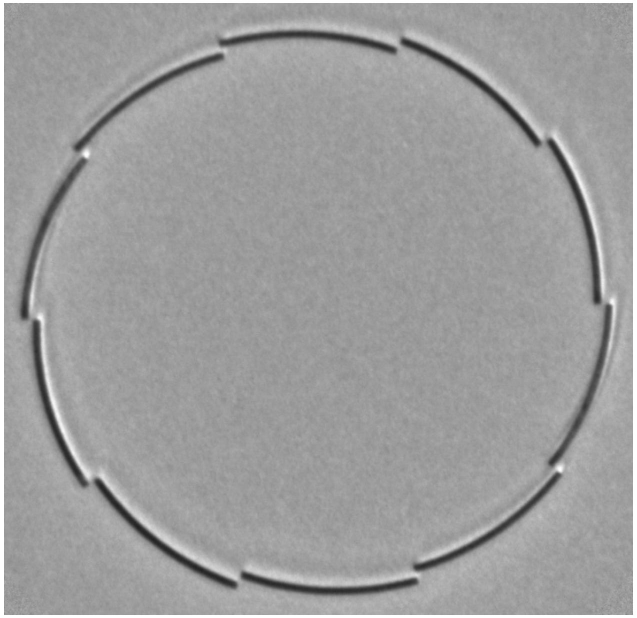
Linear $l = m + s$

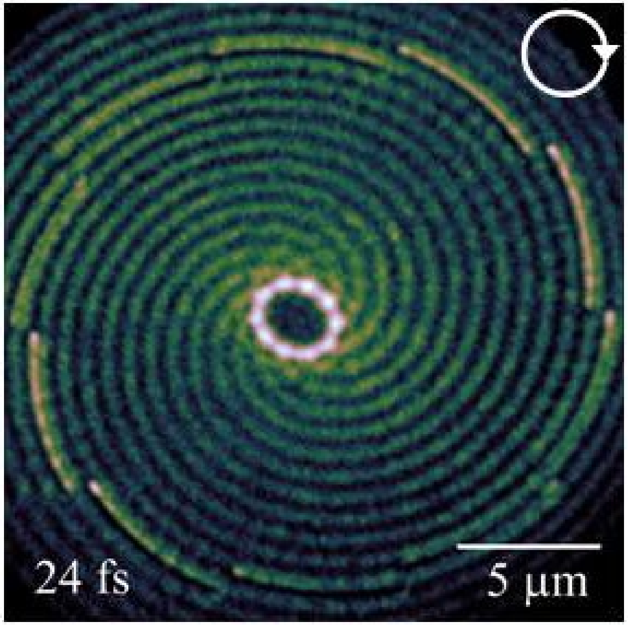
Nonlinear $l_{nl} = m + s_1 - s_2$



All optical control of OAM
delivered to the material in a
nonlinear interaction process

Optical spin-orbit coupling conversion process





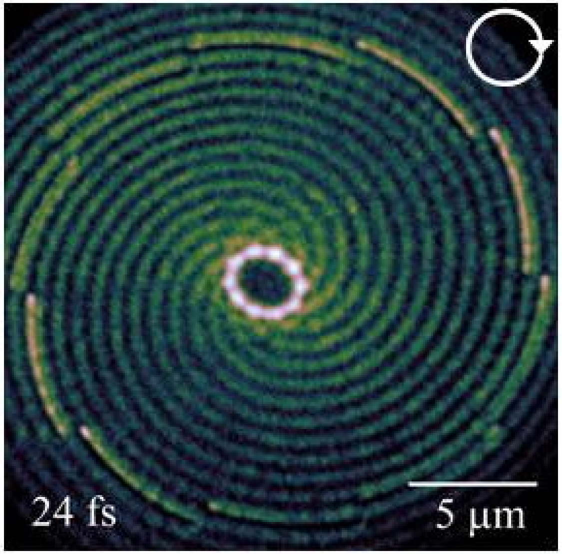
- conversion process occurs upon the interaction of the illumination with the structure
- conversion process localized to the structure boundary
- once the SPPs are launched with the proper phases by the boundary, the **topological charge** of the to-be-formed vortex is predetermined.

Mixing the Light-Spin with Plasmon-Orbit

Spin (SAM)

$$S_z = \pm\hbar$$

Orbital (OAM)

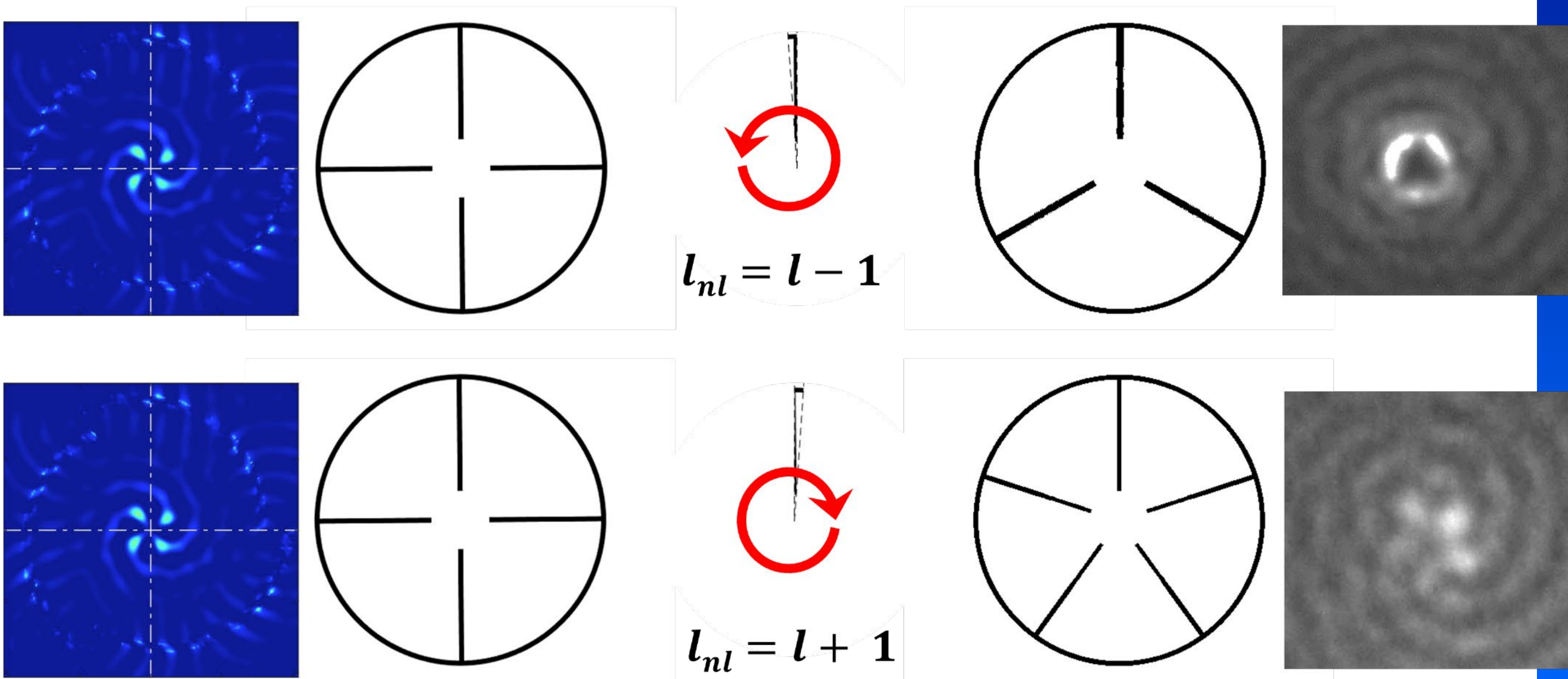


Interaction of a circularly polarized light pulse with a propagating (rotating) plasmonic vortex
Mixing of different forms of angular momentum

Measured total OAM (e.g. by 2PPE) depends on the rotating frame of the probe

Intuition rotating frame of the probe

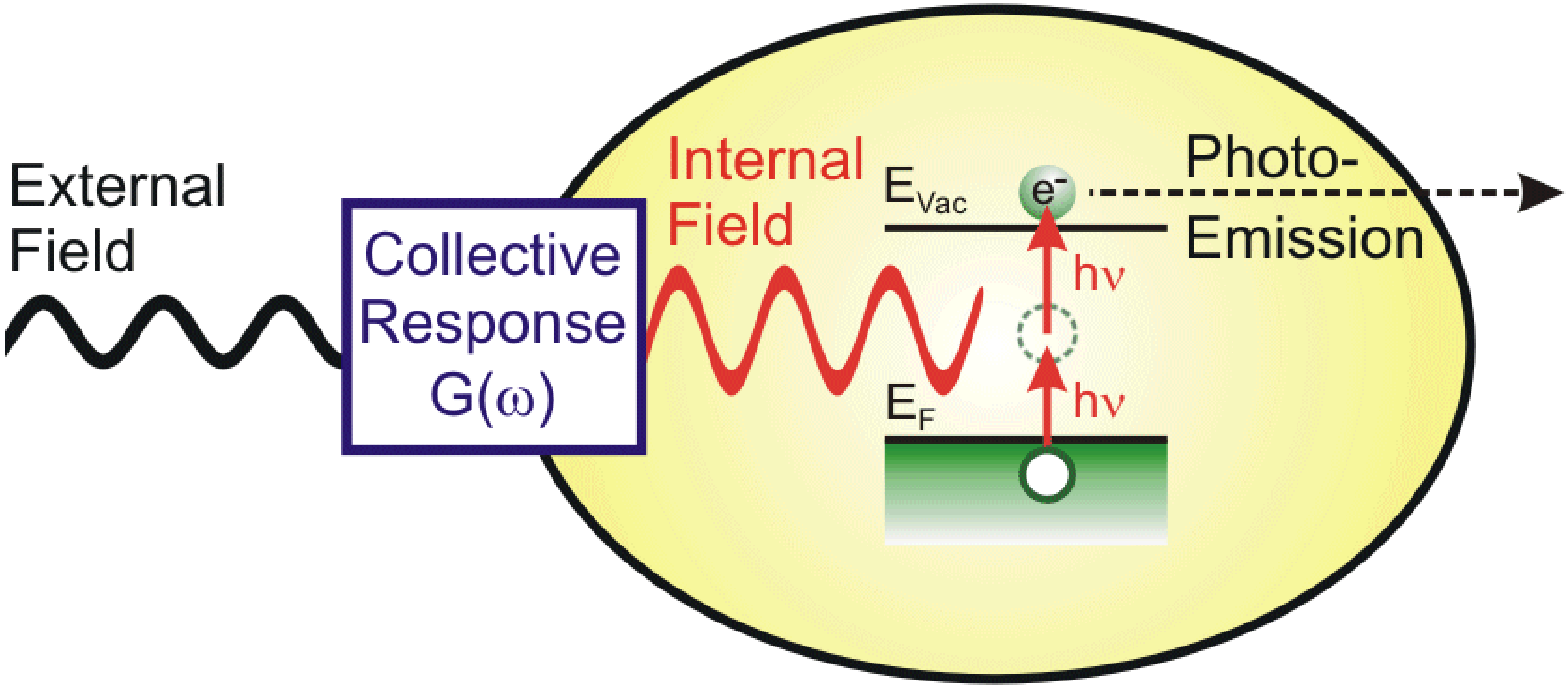
Vortex Field **Probe** **Measured**



angular momentum carried by the vortex

angular momentum carried by the light

Non-linear photoemission microscopy

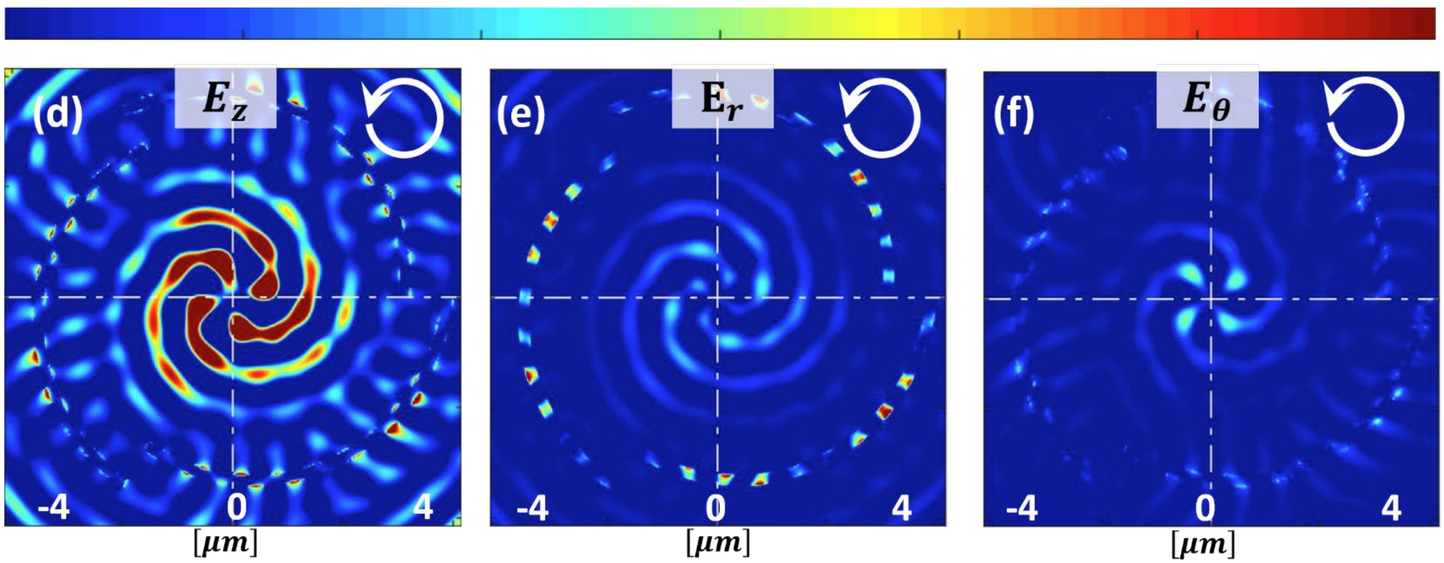


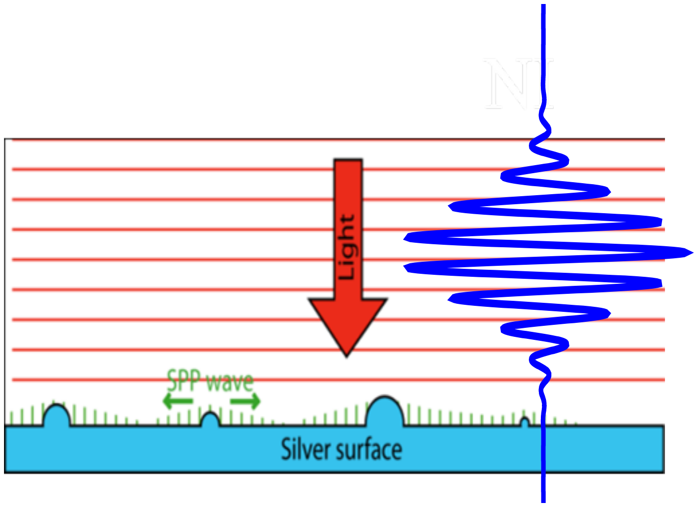
$$S_{2PPE} \propto \left(\vec{E}_{vortex} + \vec{E}_{light}\right)^4$$

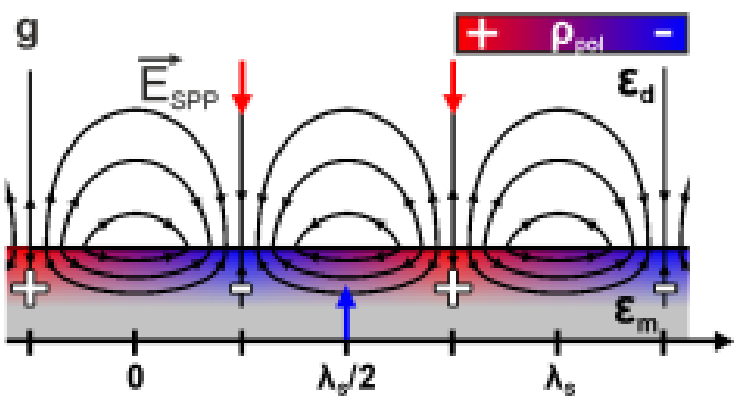
In a PEEM image, what do we really see?

Nonlinear optical spin-orbit interaction

out-of-plane component in-plane radial and azimuthal component

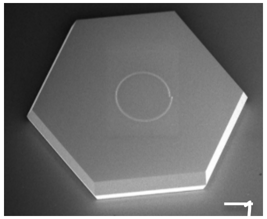






F. Meyer zu Heringdorf, Duisburg

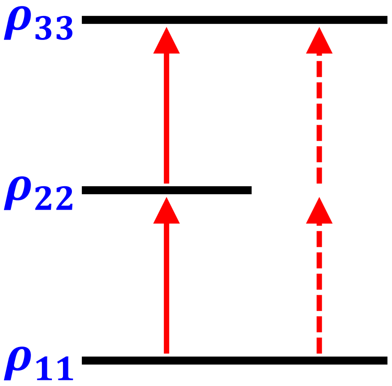
Time domain modeling of the dynamic PEEM signal



the instantaneous photoemission is given by:

$$I_{inst}(x,y,t) \propto \left[\vec{E}_{light}(x,y,t) + \vec{E}_{SPP}(x,y,t)\right]^4$$

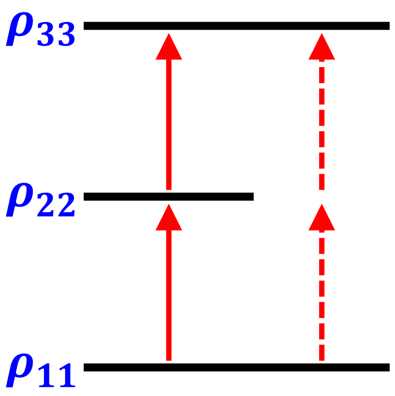
$$I_{inst} = {E_{spp,x}}^4 + {E_L}^4 + \alpha^4 {E_{spp,z}}^4 + 4{E_{spp,x}}^3 E_L + 6{E_{spp,x}}^2 {E_L}^2 + 4E_{spp,x}{E_L}^3 +$$
$$+2\cdot\alpha^2 {E_{spp,z}}^2 {E_L}^2 + 4\cdot\alpha^2 {E_{spp,z}}^2 E_{spp,x} E_L + 2\cdot\alpha^2 {E_{spp,z}}^2 {E_{spp,x}}^2$$



Mixing terms: $\Rightarrow$ $I_{int} \propto {E_{spp,x}}^i {E_L}^j$

2PPE in a three-level system

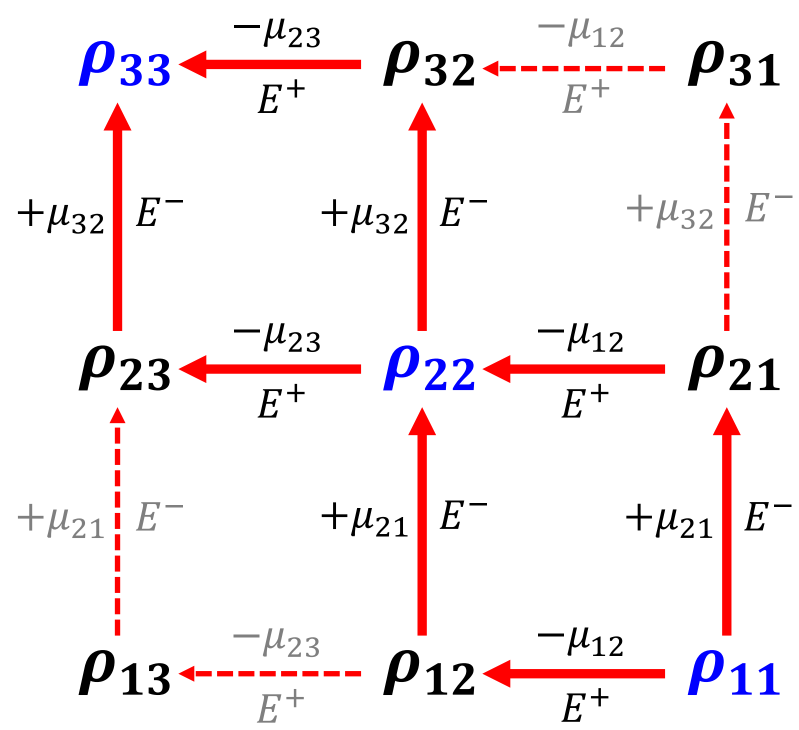
M. Merschdorf, PhD thesis (2002)



first generation of coherence
non-diagonal elements

then population of the next state

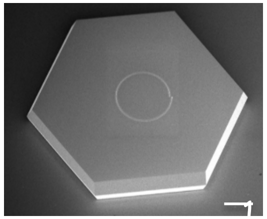
Density Matrix:



Dynamics of the system are described by the
Liouville-von Neumann equation

$$i\hbar \frac{\mathrm{d}\rho}{\mathrm{d}t} = \left[\hat{H}\,,\rho\right] + i\hbar \left.\frac{\partial \rho}{\partial t}\right|_{diss}$$

Time domain modeling of the dynamic PEEM signal



the instantaneous photoemission is given by:

$$I_{inst}(x,y,t) \propto \left[\vec{E}_{light}(x,y,t) + \vec{E}_{SPP}(x,y,t)\right]^4$$

$$I_{inst} = {E_{spp,x}}^4 + {E_L}^4 + \alpha^4 {E_{spp,z}}^4 + 4{E_{spp,x}}^3 E_L + 6{E_{spp,x}}^2 {E_L}^2 + 4E_{spp,x}{E_L}^3 +$$
$$+2\cdot\alpha^2 {E_{spp,z}}^2 {E_L}^2 + 4\cdot\alpha^2 {E_{spp,z}}^2 E_{spp,x} E_L + 2\cdot\alpha^2 {E_{spp,z}}^2 {E_{spp,x}}^2$$

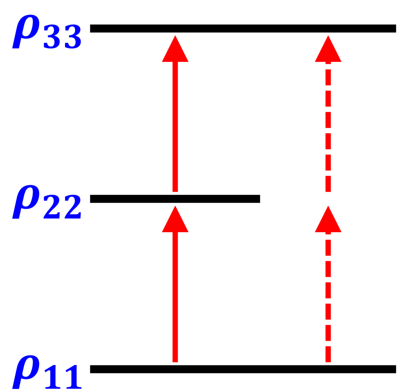
Mixing terms: $\Rightarrow$ $I_{int} \propto {E_{spp,x}}^i {E_L}^j$

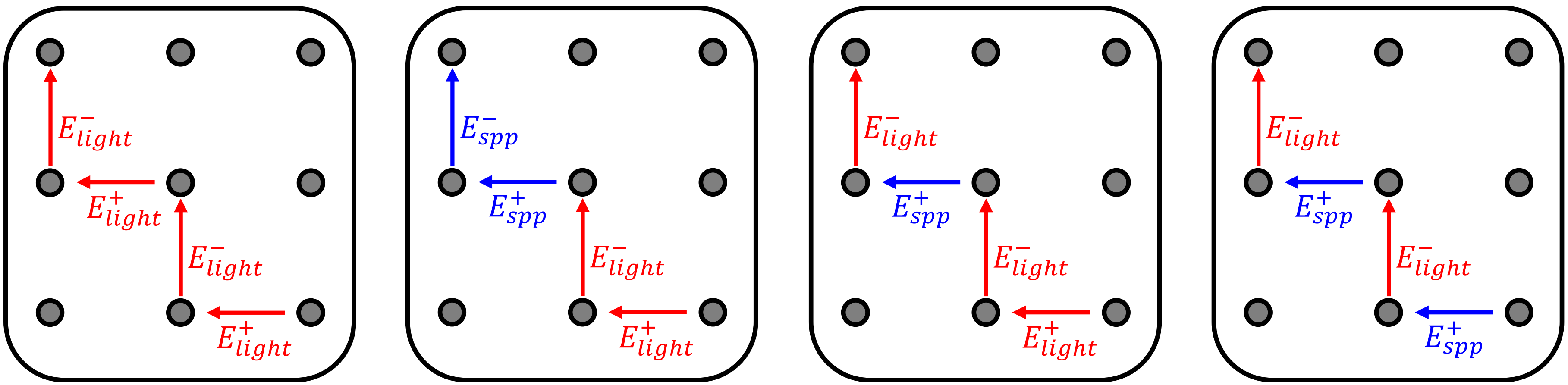
azimuthal phase dependence

Single angular momentum mixing $\propto \cos((\boldsymbol{l} - \boldsymbol{\sigma})\theta - \omega\Delta t)$ 10 lobes

Double angular momentum mixing $\propto \cos(\mathbf{2}(\boldsymbol{l} - \boldsymbol{\sigma})\theta - 2\omega\Delta t)$ 20 lobes

Quantum pathways in the density matrix

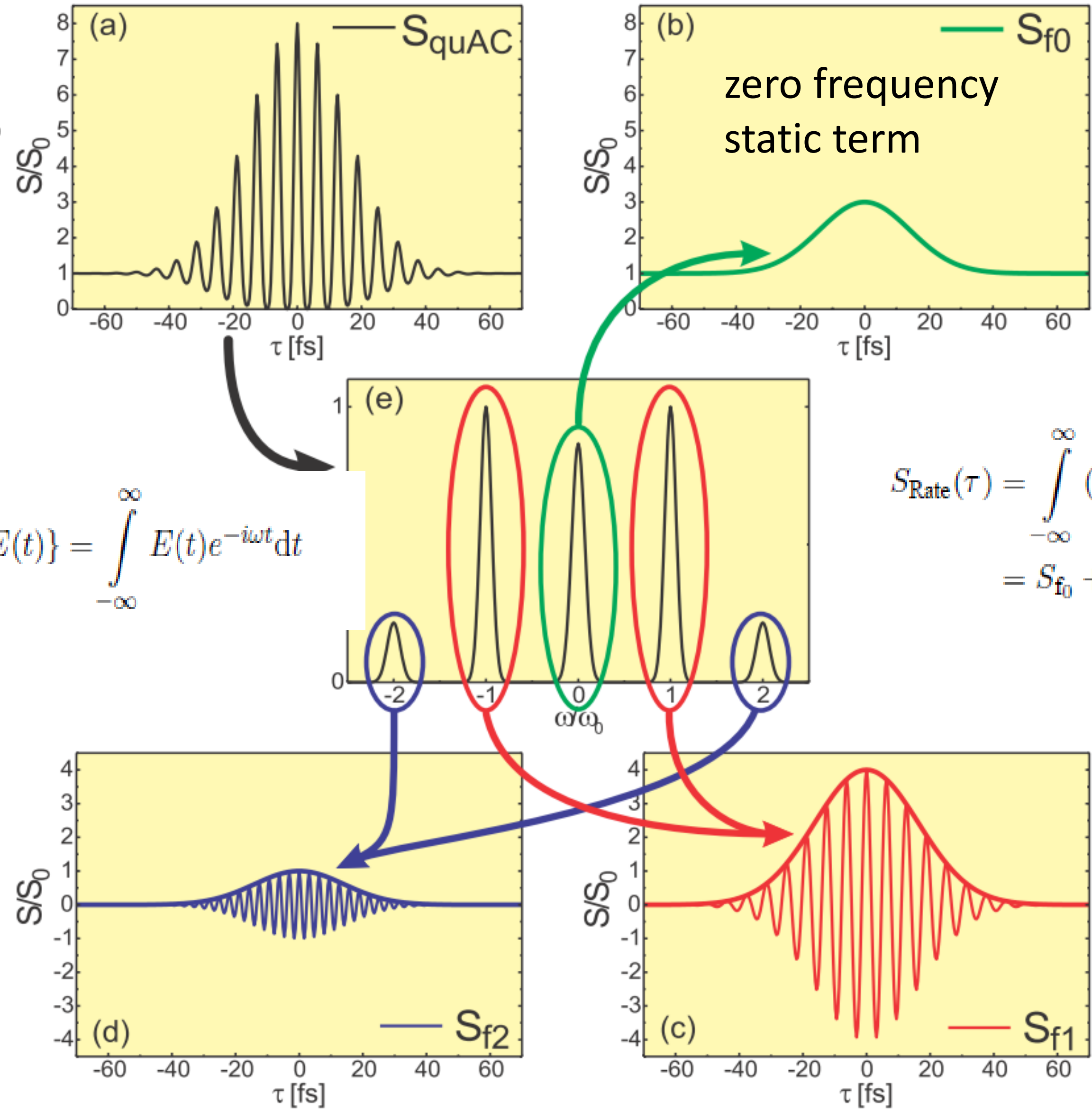




	no momentum mixing	single mixing	double mixing
		$\propto \cos((\boldsymbol{l}-\boldsymbol{\sigma})\theta)$	$\propto \cos(\mathbf{2}(\boldsymbol{l}-\boldsymbol{\sigma})\theta)$
azimuthal phase dependence		10 lobes	20 lobes

Second order two-pulse autocorrelation: Transition in the frequency domain

M. Merschdorf,
PhD thesis (2002)



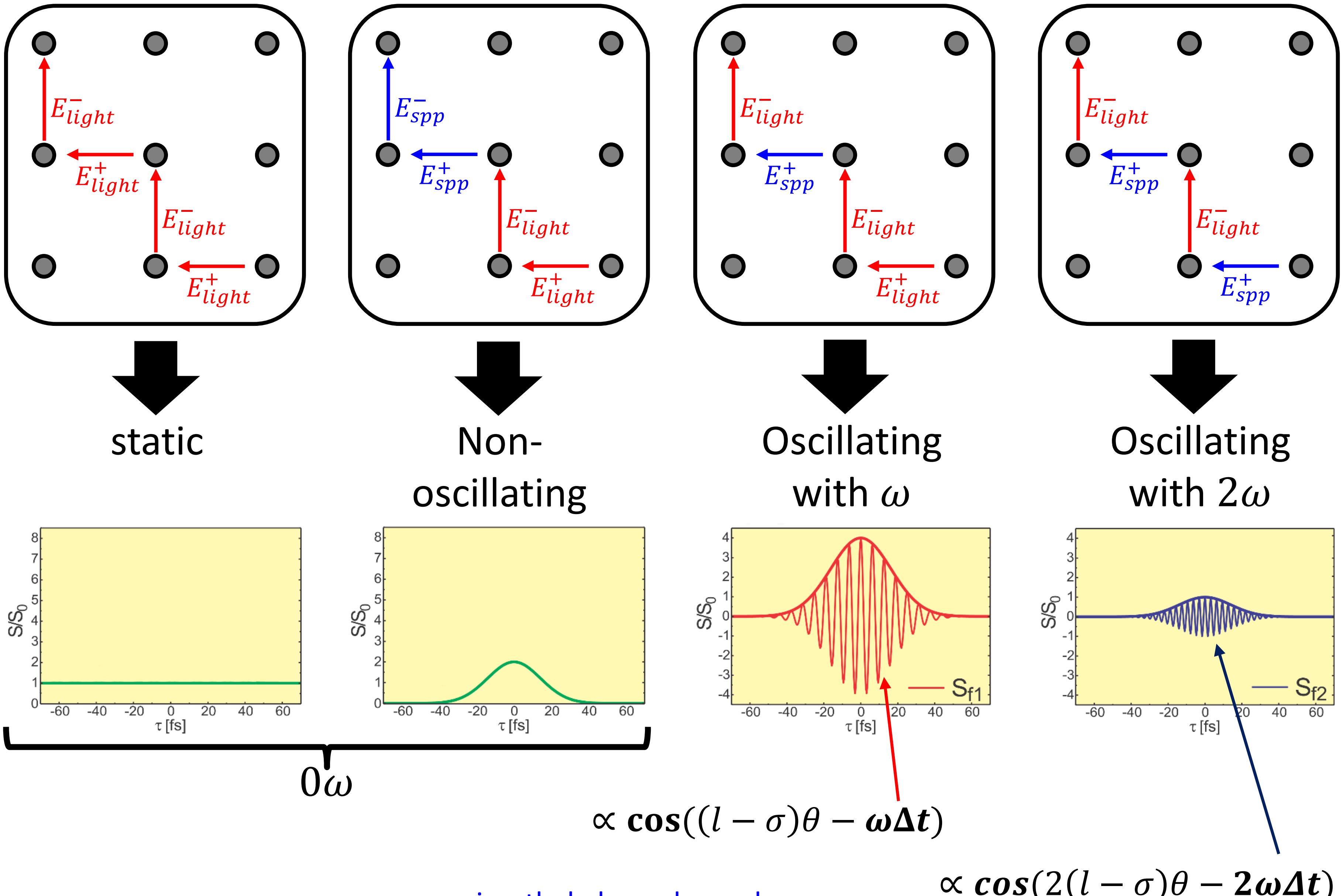
$$E(\omega) = \mathcal{F}\{E(t)\} = \int_{-\infty}^{\infty} E(t)e^{-i\omega t}\mathrm{d}t$$

$$S_{\mathrm{Rate}}(\tau) = \int_{-\infty}^{\infty} (I_{1+2} \circ G_{\mathrm{eff}})I_{1+2}\mathrm{d}t$$
$$= S_{\mathrm{f}_0} + S_{\mathrm{f}_1} + S_{\mathrm{f}_2}$$

dynamic second order term

dynamic first order term

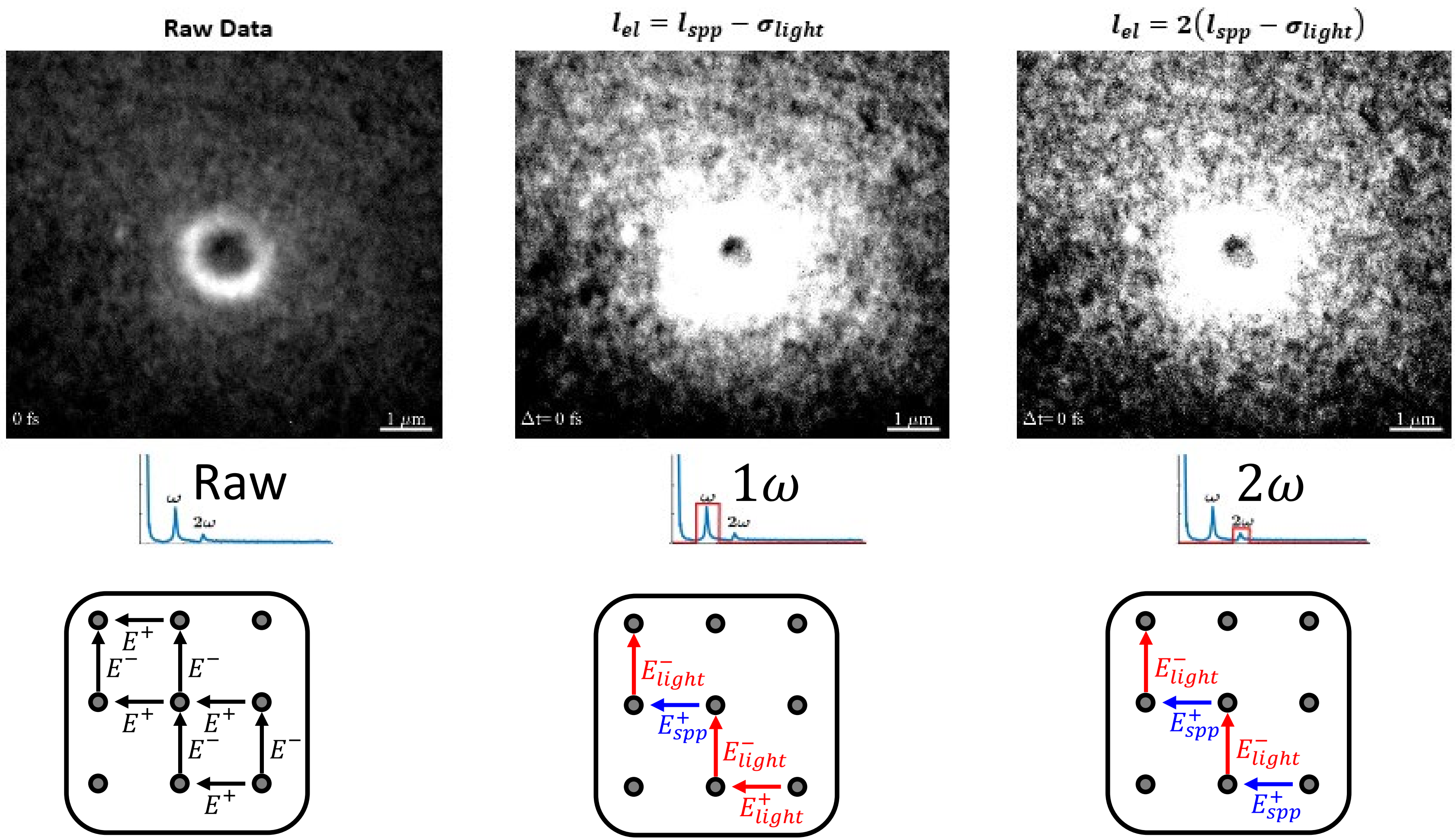
Quantum pathways in the density matrix



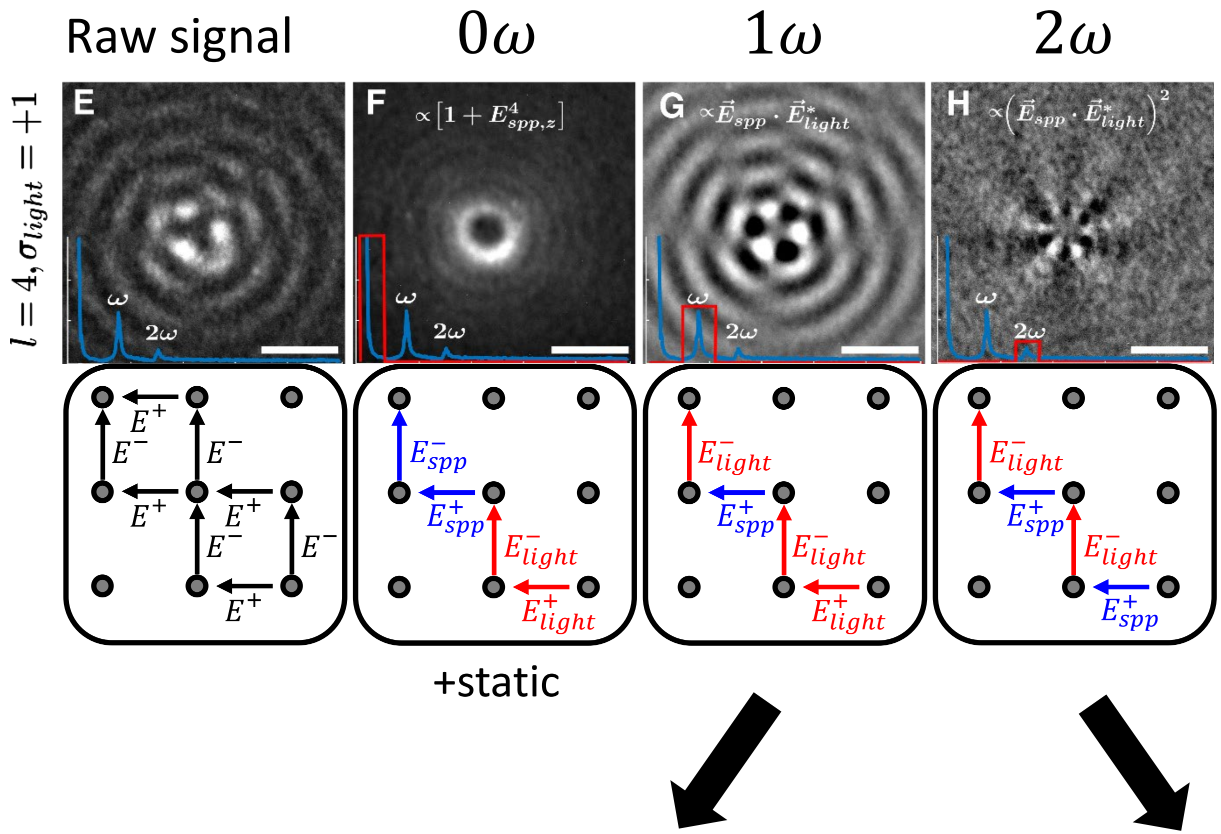
azimuthal phase dependence

Separating quantum pathways via Fourier transformation

$$l_{spp} = 4, \sigma_{light} = +1$$



Separating quantum pathways via Fourier transformation



Angular momentum mixing

$$\propto \cos((\boldsymbol{l} - \boldsymbol{\sigma})\theta - \omega\Delta t)$$

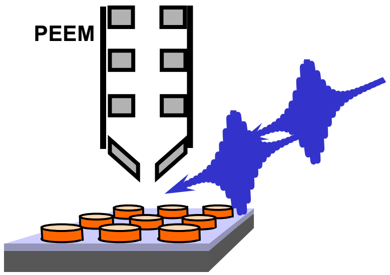
Double angular momentum mixing

$$\propto \cos(\mathbf{2}(\boldsymbol{l} - \boldsymbol{\sigma})\theta - 2\omega\Delta t)$$

Phys. Rev. X **9**, 021031 (2019)
ACS Photonics 2023, 10, 340−367

Summary

- Time-resolved interferometric PEEM technique



- Dynamics of OAM in plasmonic vortices: Real time view of a spiral phase,

- Demonstration of nonlinear optical spin-orbit conversion