Propriétés de polarisation des plasmons de surface : entre optique singulière et forces optiques

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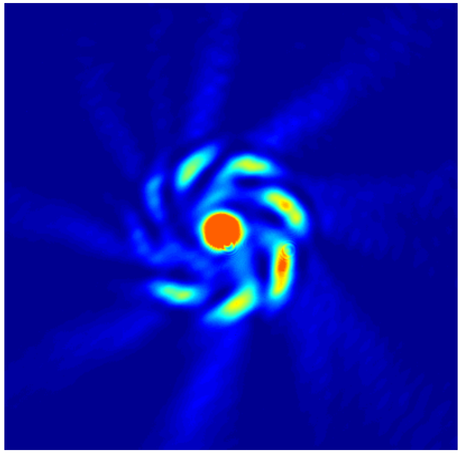


Avec : A. Canaguier-Durand, A. Cuche, A. Drezet, Y. Gorodetski

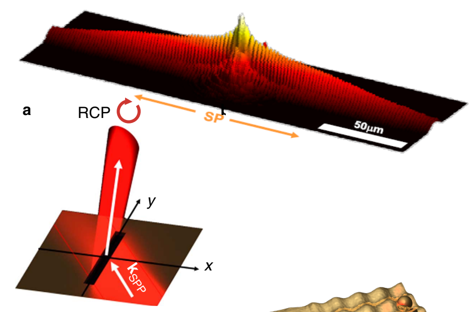
Plan

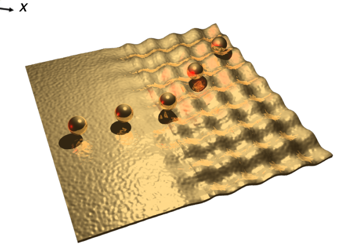
1 - Surface plasmons and polarization

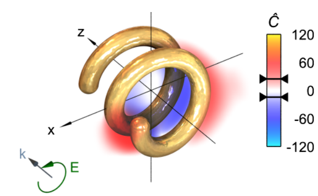
2 – Surface plasmons and spin-orbit coupling

3 – Surface plasmons and optical forces

4 – Surface plasmons and enhanced chiroptical spectroscopy



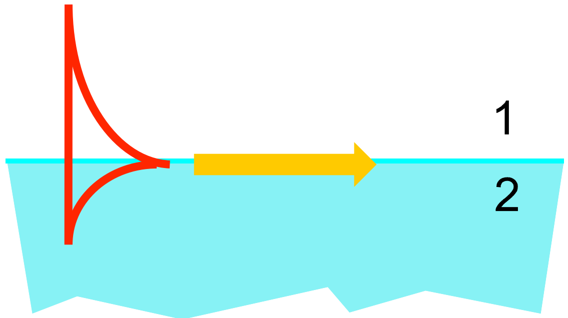


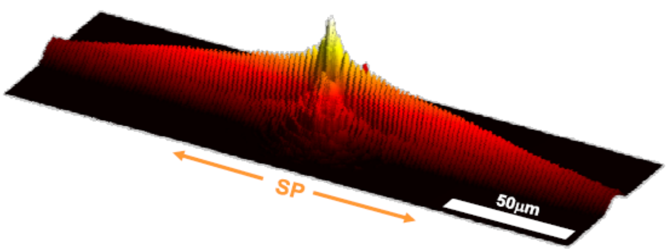


- *Spinning fields*
- *Confined energy*
- *Chirality*

Surface plasmon mode

dielectric (1) / metal (2) interface





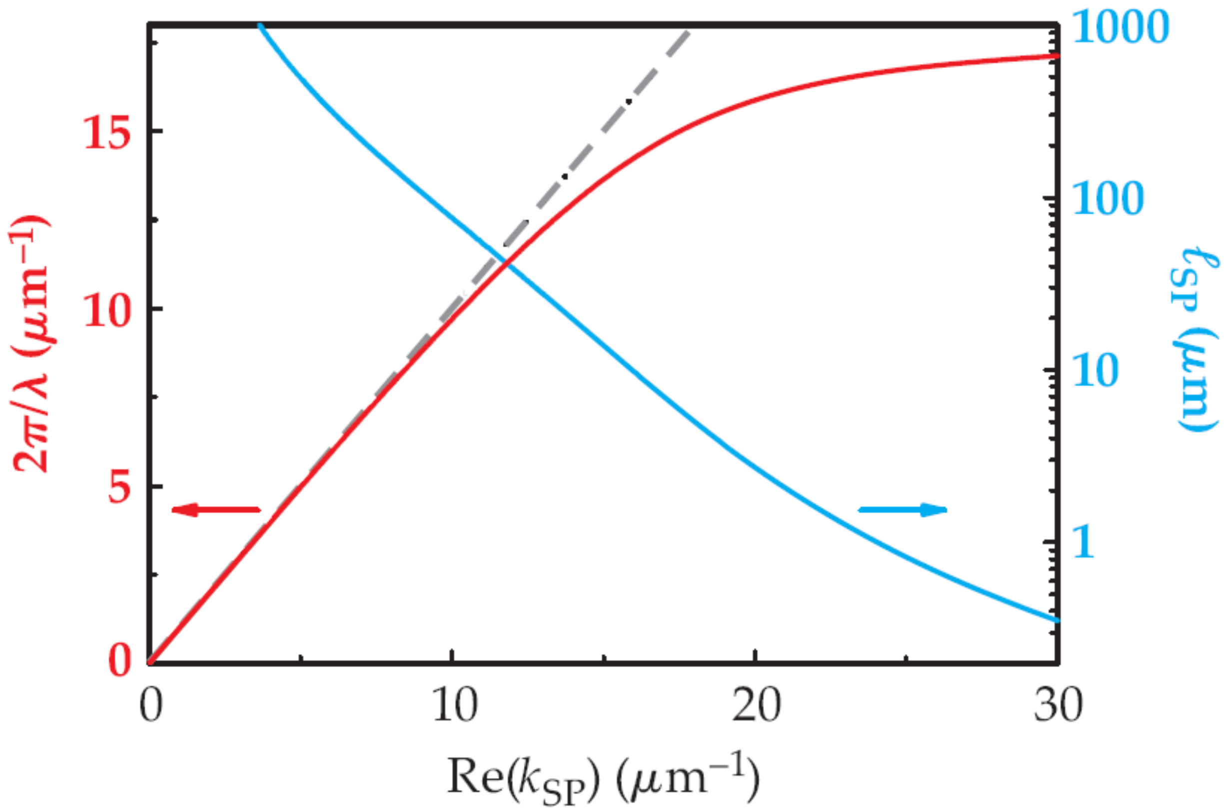
$$r = \frac{Z_1 - Z_2}{Z_1 + Z_2}$$

$$Z_1^{\mathrm{TM}} + Z_2^{\mathrm{TM}} = 0$$

$$k = \frac{\omega n_d}{c}\sqrt{\frac{\varepsilon_m}{\varepsilon_d + \varepsilon_m}}$$

- propagates along the interface (40 μm at 800nm)
- confines the field at the interface
- highly sensitive to *n*

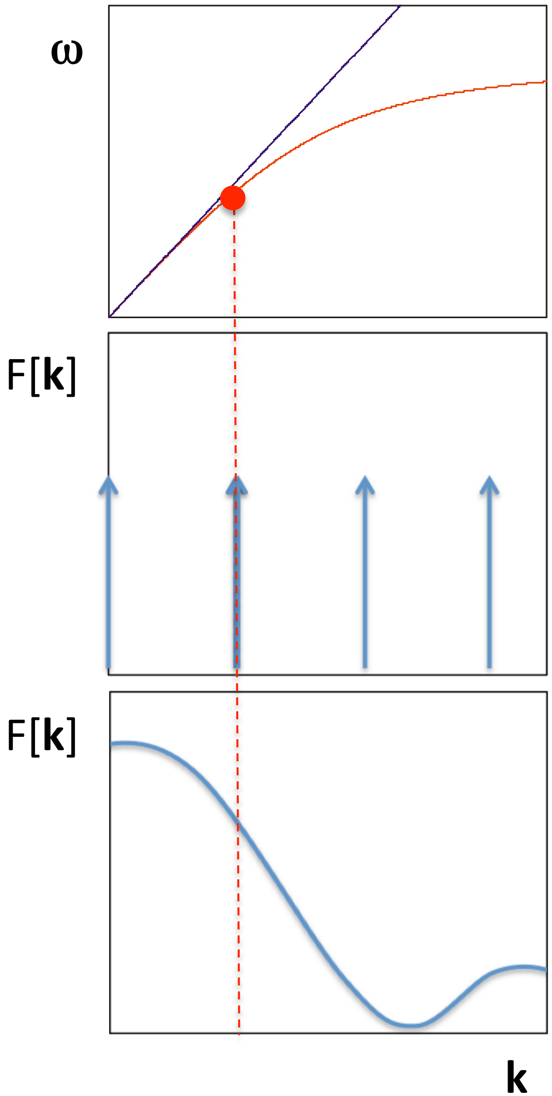
Dispersion relation

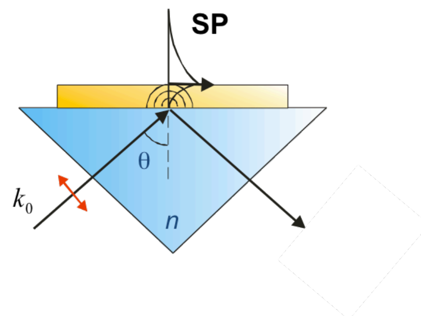


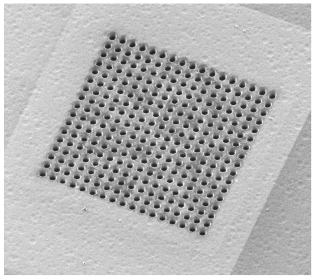
□ Well-defined mode: $\ell_{\mathrm{SP}} > \lambda_{\mathrm{SP}}$

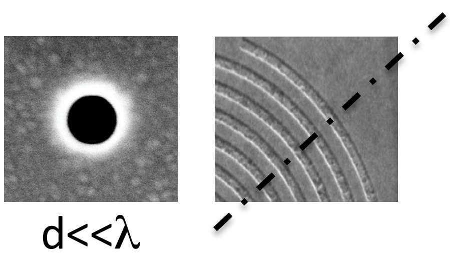
$$|\mathrm{Re}[k]| > \mathrm{Im}[k] > 0 \qquad |\mathrm{Re}[\varepsilon_m]| > \varepsilon_d$$

Coupling schemes







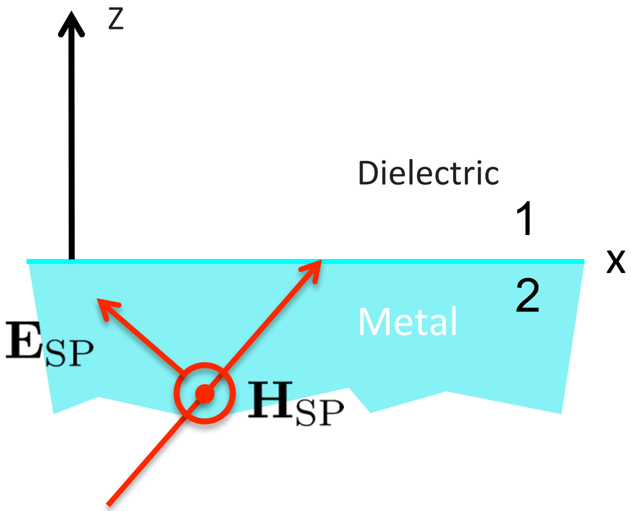


- 70-90s: SP optics, thin films spectroscopy, refractive index sensing, etc.
- 00s: EOT, plasmonic crystals, etc.
- sub-wavelength apertures and *extended* structures

Surface plasmon field

$$\tilde{u} = \frac{c}{\omega n_d} u$$

- SP field: TM-polarized



$$\mathbf{H}^d_{\rm SP} = H_0[0,1,0]e^{ikx}e^{iq^d z}e^{-i\omega t}$$

$$\mathbf{H}^m_{\rm SP} = H_0[0,1,0]e^{ikx}e^{-iq^m z}e^{-i\omega t}$$

$$k = k' + ik'' \qquad k'' > 0$$

$$q^{d,m} = \sqrt{\varepsilon_{d,m}(\omega/c)^2 - k^2}$$

$$= (q^{d,m})' + i(q^{d,m})'' \qquad (q^{d,m})'' > 0$$

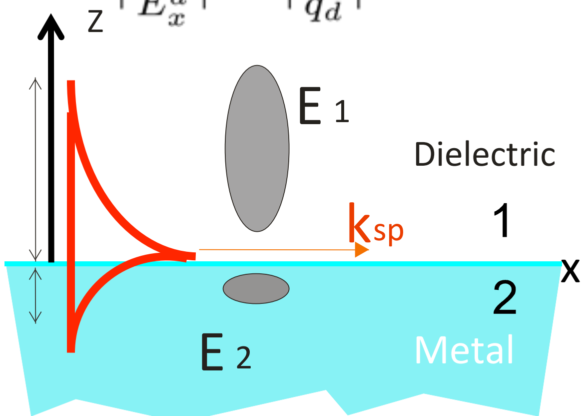
$$\frac{\partial (q^{d,m})''}{\partial k'} < 0$$

Surface plasmon field

$$\tilde{u} = \frac{c}{\omega n_d} u$$

$$\mathbf{E}_{\mathrm{SP}} = E_0[\tilde{q}, 0, -\tilde{k}] e^{ikx} e^{iqz}$$

$$|\frac{E_z}{E_x^d}| = |\frac{\tilde{k}}{\tilde{q}_d}| > 1$$



$$|\frac{E_z}{E_x^m}| = |\frac{\tilde{k}}{\tilde{q}_m}| < 1$$

□ Scales

Attenuation	$\ell_i = 1/2\mathrm{Im}[k, q_i]$
longitudinal $k = \frac{\omega n}{c}\sqrt{\frac{\varepsilon_m}{\varepsilon_m + \varepsilon_d}}$	λ=800nm, Au/air $\ell_{\mathrm{SP}} \sim$ 40 μm
transverse $q_i = \frac{\omega}{c}\sqrt{\frac{\varepsilon_i^2}{\varepsilon_m + \varepsilon_d}}$	$\ell_d \propto \lambda\sqrt{\varepsilon_m} \sim$ 200 nm $\ell_m \propto \frac{\lambda}{\sqrt{\varepsilon_m}} \sim$ 15 nm

$<< \lambda$

Field ellipticity

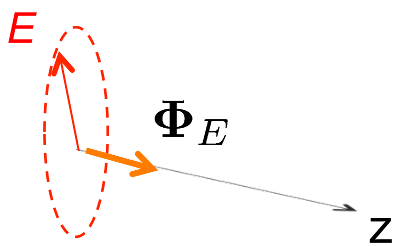
- Elliptical polarization

Complex field $$\mathbf{E}(\mathbf{r},t) = \mathbf{E}_0(\mathbf{r})e^{-i\omega t} = [\mathbf{A}(\mathbf{r}) + i\mathbf{B}(\mathbf{r})]\, e^{-i\omega t}$$

Real field $$\mathcal{E}(\mathbf{r},t) = \mathbf{A}(\mathbf{r})\cos\omega t + \mathbf{B}(\mathbf{r})\sin\omega t$$

- A measure for the ellipticity (Berry, 2000)

$$\mathbf{\Phi}_E(\mathbf{r}) = \mathbf{A}(\mathbf{r}) \times \mathbf{B}(\mathbf{r})$$
$$= -\tfrac{1}{2}\mathrm{Im}\,[\mathbf{E} \times \mathbf{E}_0^\star]$$



A connection with the spin density

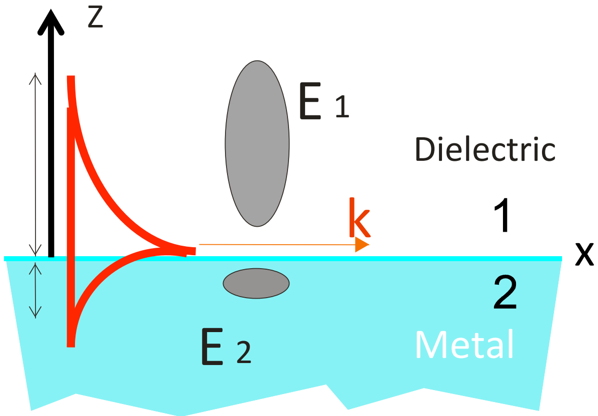
$$\mathbf{\Phi}_E = \mathcal{E} \times \dot{\mathcal{E}}/\omega = \langle||\mathcal{E}||^2\rangle_T\ \mathbf{S}_E$$

$$\mathbf{E}(\mathbf{r},t) = \mathcal{F}(\mathbf{r}_\perp)[\mathbf{x} + i\mathbf{y}]e^{-i\omega t}$$

(dire que cette relation clair à partir de forces)

Surface plasmons as spinning near fields

□ TM-polarized spinning field



$$\mathbf{H}_{\text{SP}} = H_0[0, 1, 0]e^{ikx}e^{iqz}$$

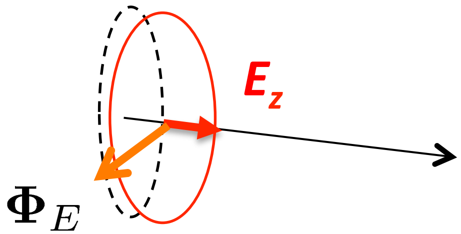
$$\mathbf{E}_{\text{SP}} = E_0[\tilde{q}, 0, -\tilde{k}]e^{ikx}e^{iqz}$$

Electric field ellipticity

$$\mathbf{\Phi}_E = \mathcal{E} \times \dot{\mathcal{E}}/\omega$$

$$\mathbf{\Phi}_E(x, z) = 2|E_0|^2 e^{-2k''x} e^{-q''z}[0, \tilde{q}'\tilde{k}'' - \tilde{k}'\tilde{q}'', 0]$$

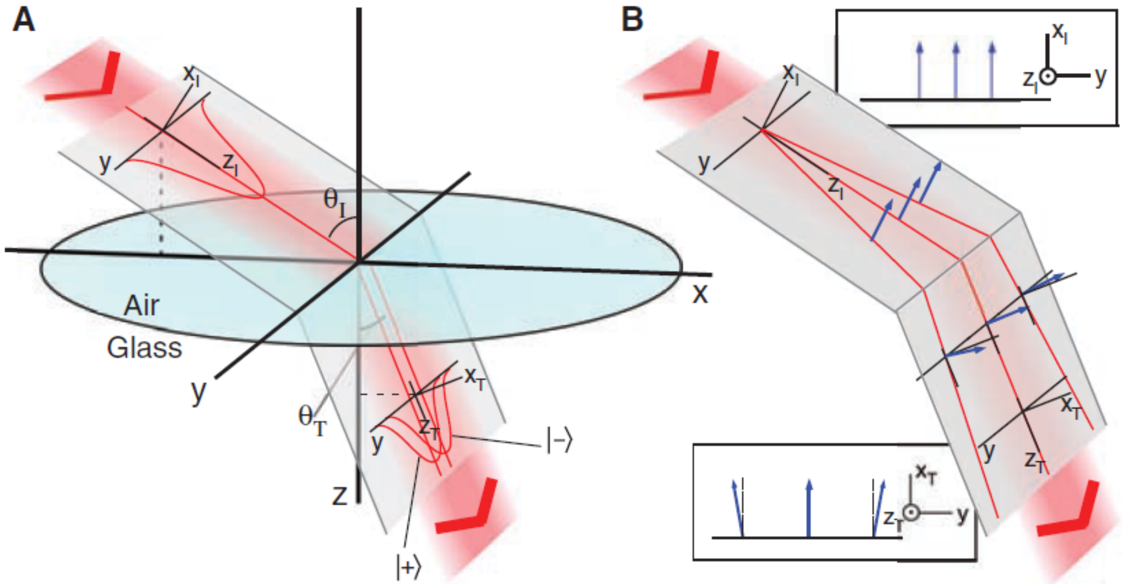
Longitudinal electric field component



Meridional plane of polarization

Spin-orbit coupling: Spin Hall Effect of Light

Hosten and Kwiat, Science ***319****, 787 (2008)*



Input state

$$|k_y\rangle|H\rangle$$

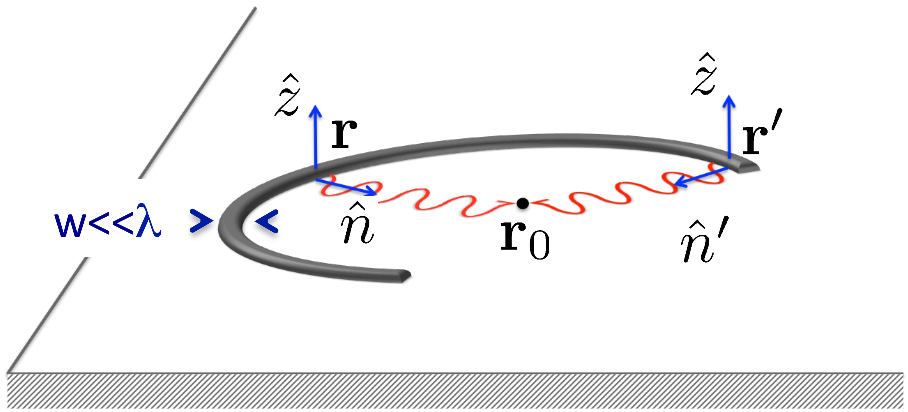
$$|\pm\rangle = \frac{1}{\sqrt{2}}\left(|H\rangle \pm i|V\rangle\right)$$

$$|k_y\rangle\left(|H\rangle + \delta k_y|V\rangle\right) = \frac{1}{\sqrt{2}}|k_y\rangle\left(e^{-ik_y\delta}|+\rangle + e^{ik_y\delta}|-\rangle\right)$$

Output state

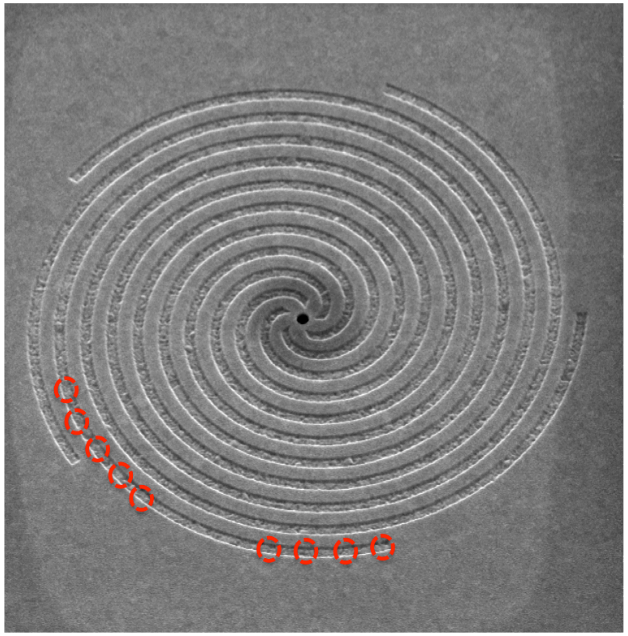
Spin-orbit coupling and surface plasmons

- Source-accompanying local frame

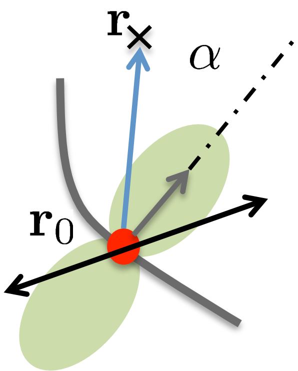


- Space variant polarization states
- Geometric phases
- Spin-orbit coupling

Huygens-Fresnel modeling



$$\mathbf{E}_{\rm dip}(\mathbf{r}) = \frac{e^{ik_{\rm SP}|\mathbf{r}-\mathbf{r}_0|}}{\sqrt{|\mathbf{r}-\mathbf{r}_0|}} \cos\alpha\,(\mathbf{E}_{\rm in}\cdot\mathbf{n})\,\mathbf{n}$$

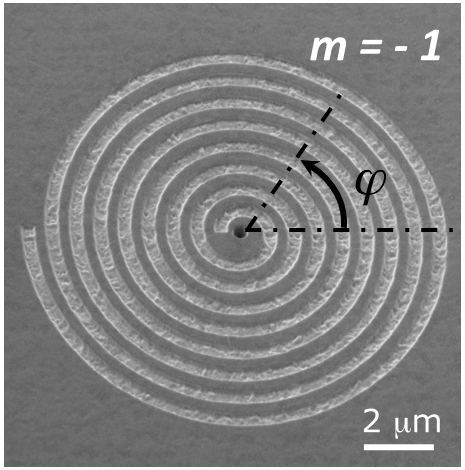


$$\mathbf{E}_{\rm SP}(\mathbf{r}) = \int_S {\rm d}s \mathbf{E}_{\rm dip}(s)$$

S – curvilign coordinate

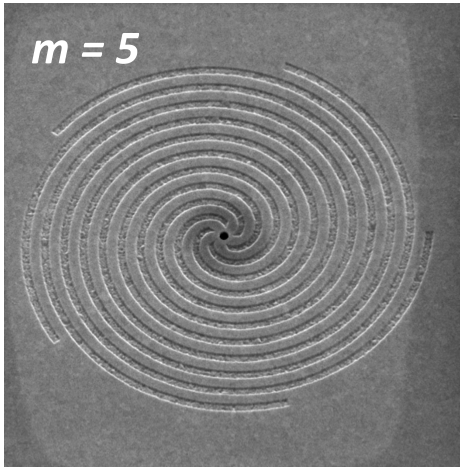
See: A. Drezet et al., J. Appl. Phys. ***115****, 093105 (2014)*

Spin-orbit coupling and chiral plasmons



$$\rho_n = (n\lambda_{\mathrm{SP}} + m\varphi\lambda_{\mathrm{SP}}/2\pi)\hat{\rho}$$

Assume radial regions $\rho_n >> \rho_0$

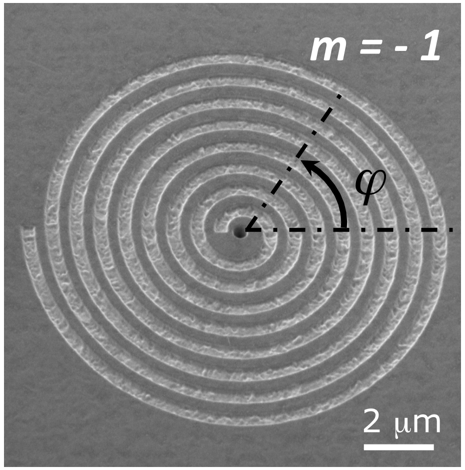


$$\mathbf{E}_{\mathrm{SP}}(\rho_0) \propto \int_0^{2\pi} \mathrm{d}\phi e^{im\phi} e^{ik_{\mathrm{SP}}\rho_0 \cos\phi} \left[\mathbf{E}_{\mathrm{in}} \cdot \hat{\rho}\right] \hat{\rho}$$

dynamic phase *geometric phase*

OAM *Spin-orbit coupling*

Spin-orbit coupling and chiral plasmons



$$\rho_n = (n\lambda_{SP} + m\varphi\lambda_{SP}/2\pi)\hat{\rho}$$

$$\mathbf{E}_{\mathrm{SP}}(\rho_0) \propto \int_0^{2\pi} \mathrm{d}\phi e^{im\phi} e^{ik_{\mathrm{SP}}\rho_0 \cos\phi} \left[\mathbf{E}_{\mathrm{in}} \cdot \hat{\rho}\right] \hat{\rho}$$

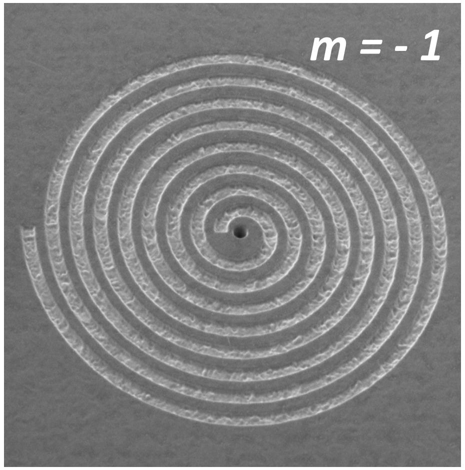
- Spin-orbit coupling

$$\mathbf{E}_{\mathrm{in}} \propto \hat{\sigma}_{\pm} = \left(\hat{\rho} \pm \hat{\phi}\right) e^{\pm i\phi}/\sqrt{2}$$

$$\mathbf{E}_{\mathrm{SP}} \propto J_{m\pm1}(k_{\mathrm{SP}}\rho) e^{i(m\pm1)\varphi}$$

For instance:
*Ohno & Miyanishi Opt. X **14**, 6285 (2006)*
*Hasman et al., Nano Letters **9**, 3016 (2009)*

Chiral plasmons for singular optics



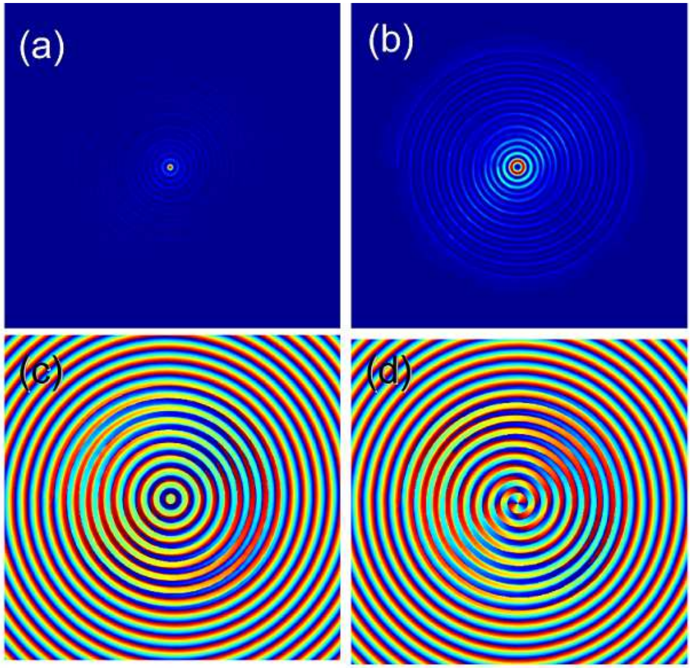
$$\mathbf{E}_{\mathrm{SP}} \propto J_{m\pm1}(k_{\mathrm{SP}}\rho)e^{i(m\pm1)\varphi}$$

Generating Bessel beams with fixed OAM

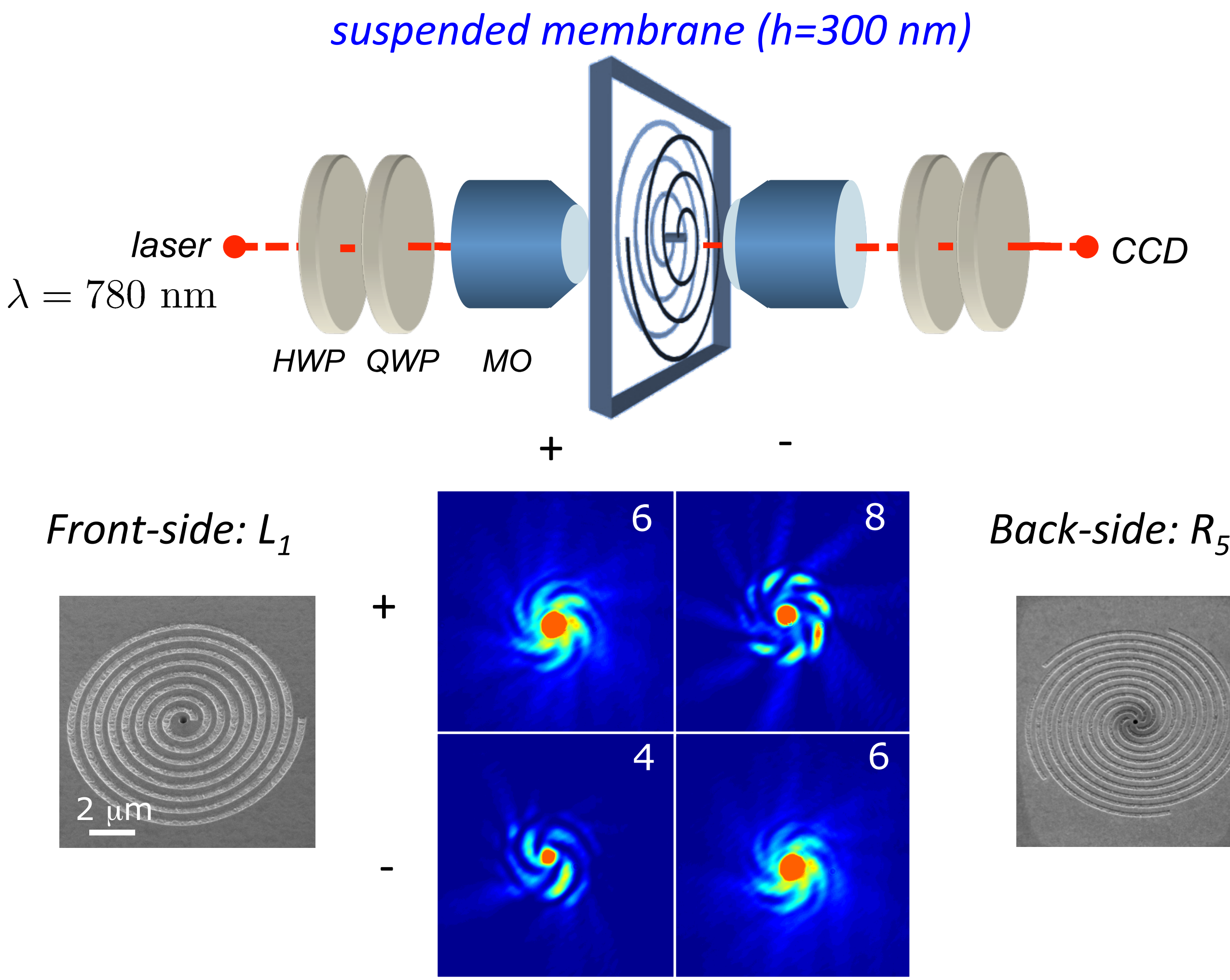
$$\ell_{\mathrm{OAM}} = m \pm 1$$

$$\hat{\sigma}_{+} \qquad \hat{\sigma}_{-}$$

$$m = -1$$

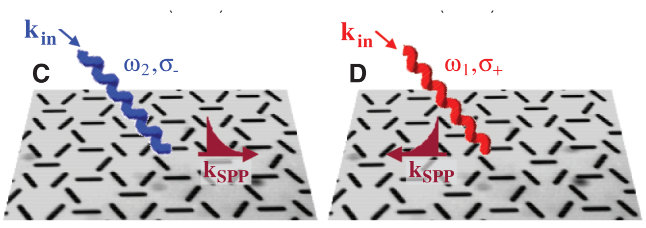


OAM transfer from near-field chirality



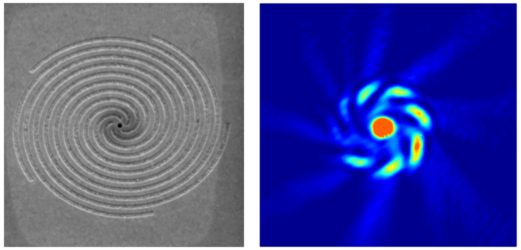
Gorodetski *et al.*, PRL ***110***, 203906 (2013)

Spin-orbit coupling in nano optics

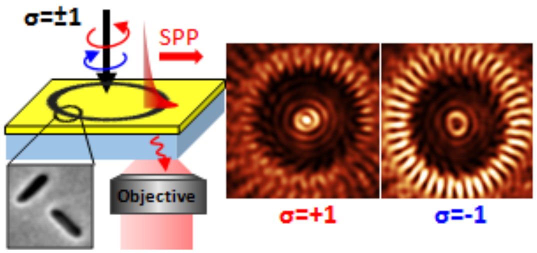


Hasman, Technion

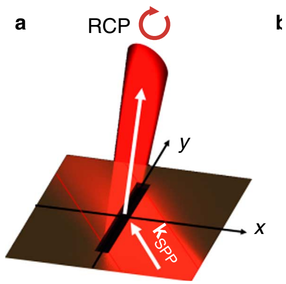
Capasso, Harvard



ISIS, Strasbourg



Drezet, Néel

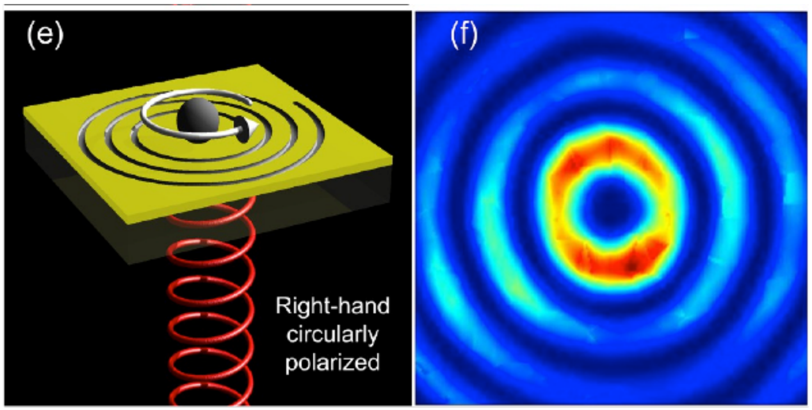


Zayats, King's college

Review: Bliokh, Rodriguez-Fortuno, F. Nori, and A.V. Zayats
Nature Photon. (2016)

Plasmonic vortices and orbiting motions

- Trapping microparticles at primary rings of plasmonic vortices



- Orbiting motions

*See: Tsai et al., Nano Letters **14**, 547 (2014)*

Optical force and torque on an electric dipole

- Lorentz law (real $(\mathcal{E},\mathcal{H})$ fields)

$$\mathbf{F} = (\mathcal{P}\cdot\nabla)\mathcal{E} + \mu_0\dot{\mathcal{P}}\times\mathcal{H}$$

$$\boldsymbol{\Gamma} = \mathcal{P}\times\mathcal{E}$$

$$\mathcal{E} = \mathrm{Re}[\mathbf{E}_0(\mathbf{r})e^{-i\omega t}]$$

$$\mathcal{P} = \mathrm{Re}[\mathbf{p}_0(\mathbf{r})e^{-i\omega t}]$$

$$\mathbf{p}_0(\mathbf{r}) = n^2\alpha\mathbf{E}_0(\mathbf{r})$$

- Time-averaged force

$$\langle\mathbf{F}\rangle_T = \frac{n^2}{2}\mathrm{Re}[\alpha\mathbf{f}_0]$$

$$\mathbf{f}_0 = \sum_j E_j\nabla E_j^*$$

$$\mathbf{F}_{\mathrm{reactive}} = \frac{n^2}{2}\mathrm{Re}[\alpha]\mathrm{Re}[\mathbf{f}_0]$$

$$\mathbf{F}_{\mathrm{dissipative}} = -\frac{n^2}{2}\mathrm{Im}[\alpha]\mathrm{Im}[\mathbf{f}_0]$$

- linear polarization $\mathbf{E}_0(\mathbf{r}) = \rho(\mathbf{r})e^{i\phi(\mathbf{r})}\hat{\mathbf{u}}$

$$\mathbf{f}_0 = \rho\nabla\rho - i\rho^2\nabla\phi$$

- general polarization case $E_0^j = \rho^j e^{i\phi^j}$

$$\mathrm{Im}[\mathbf{f}_0] = -\sum_j \rho_j^2\,\nabla\phi_j$$

Stenholm, RMP (1986)
Hemmerich & Hänsch, PRL (1992)

Radiation pressure and orbital energy flow

- Time-averaged Poynting vector

$$\mathbf{\Pi} = \langle \mathcal{E} \times \mathcal{H} \rangle_T$$
$$= \mathbf{\Pi}_O + \mathbf{\Pi}_S$$

$$\mathbf{\Pi}_O = -\frac{1}{2\omega\mu_0}\mathrm{Im}[\mathbf{f}_0]$$

$$\mathbf{\Pi}_S = \frac{1}{2\omega\mu_0}\nabla \times \mathbf{\Phi}_E$$

Electric ellipticity $\mathbf{\Phi}_E = \mathcal{E} \times \dot{\mathcal{E}}/\omega$

Mechanical energy transfers through dissipation

- Time-averaged radiation pressure

$$\mathbf{F}_{\text{dissipative}} = n^2\omega\mu_0\mathrm{Im}[\alpha]\left(\mathbf{\Pi} - \frac{\nabla \times \mathbf{\Phi}_E}{2\omega\mu_0}\right)$$
$$= n^2\omega\mu_0\mathrm{Im}[\alpha]\mathbf{\Pi}_O$$

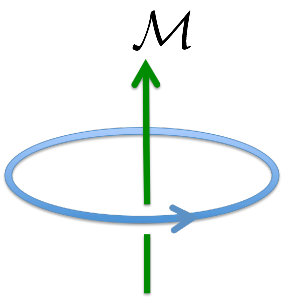
- Time-independent torque

$$\mathbf{\Gamma} = n^2\mathrm{Im}[\alpha]\mathbf{\Phi}_E$$

Canaguier et al., PRA ***88****, 033831 (2013)*

« Electric-magnetic democraty » (M.V. Berry)

- Lorentz law for a magnetic dipole



$$\mathbf{F} = (\mathcal{M}\cdot\nabla)\mathcal{H} - \varepsilon_0 \dot{\mathcal{M}} \times \mathcal{E}$$

$$\boldsymbol{\Gamma} = \mathcal{M} \times \mathcal{H}$$

- Harmonic fields and induced dipole

$$\mathbf{m}_0(\mathbf{r}) = \mu\beta\mathbf{H}_0(\mathbf{r})$$

$$\beta/\alpha \sim (ka)^2 \ll 1$$

- Dual symmetric expressions

$$\boldsymbol{\Phi} = \frac{\omega}{2}\left(\varepsilon\boldsymbol{\Phi}_E + \mu\boldsymbol{\Phi}_H\right)$$

$$\boldsymbol{\Pi}_O = \frac{1}{2}\left(\boldsymbol{\Pi}_O^{(E)} + \boldsymbol{\Pi}_O^{(H)}\right)$$

$$\boldsymbol{\Pi}_S = \frac{1}{2}\left(\boldsymbol{\Pi}_S^{(E)} + \boldsymbol{\Pi}_S^{(H)}\right) = \frac{1}{4\omega\varepsilon\mu}\nabla\times\boldsymbol{\Phi}$$

Spin and orbital angular momentum densities

Berry, J. Opt. A (2009)
Bliokh, et al. NJP (2014)

- Orbital angular momentum (time ave.)

$$\mathbf{L} = \mathbf{r} \times \mathbf{\Pi}_O$$

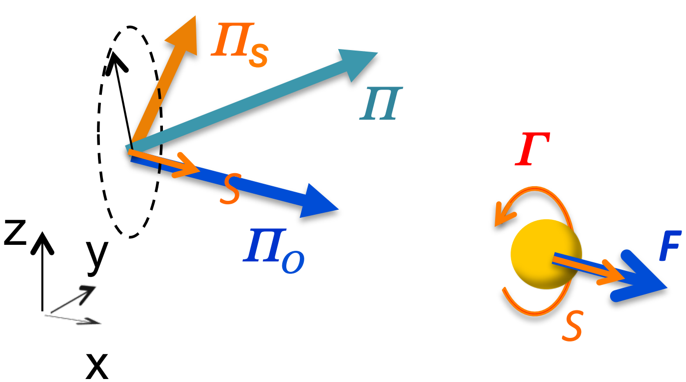
- *extrinsic*
- *transverse w.r.t. wave momentum*

- Spin angular momentum

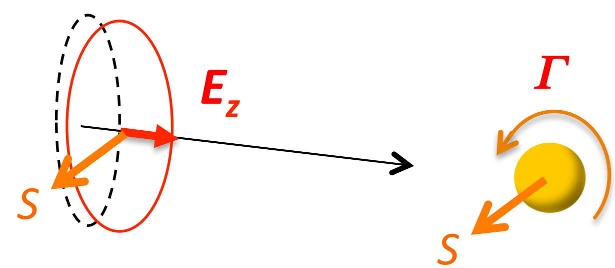
$$\mathbf{S} = \frac{c^2}{\omega^2}\mathbf{\Phi}$$

- *intrinsic*
- *no specified direction w.r.t. wave momentum*

- Transverse polarization

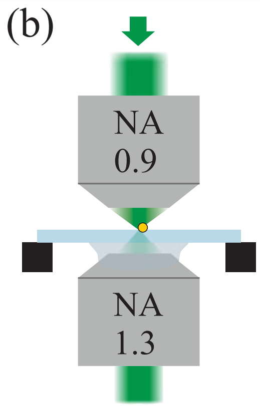


- Longitudinal electric field component *(TM polarized)*



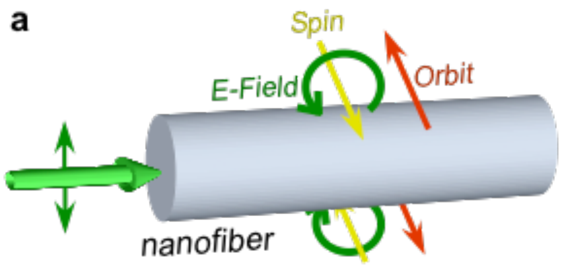
Transverse spin densities and structures light fields

- Non-paraxial fields



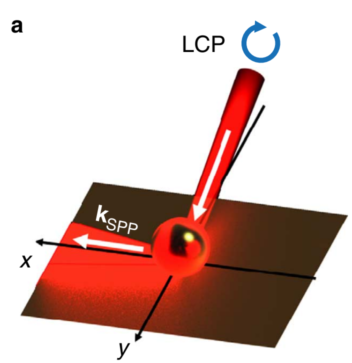
Neugbauer et al., PRL 114, 063901 (2015)

- Evanescent waves



Petersen et al., Science 346, 67 (2014)

- Surface plasmon modes



O'Connor et al., Nature Comm. 5, 5327 (2014)

Surface plasmons as spinning near fields

□ Surface plasmons

$$\mathbf{H}_{\text{SP}} = H_0[0,1,0]e^{ikx}e^{iqz}$$

$$\mathbf{E}_{\text{SP}} = E_0[\tilde{q},0,-\tilde{k}]e^{ikx}e^{iqz}$$

$$\mathbf{\Phi}_{\text{SP}} = \mathcal{E}_{\text{SP}} \times \dot{\mathcal{E}}_{\text{SP}}/\omega$$

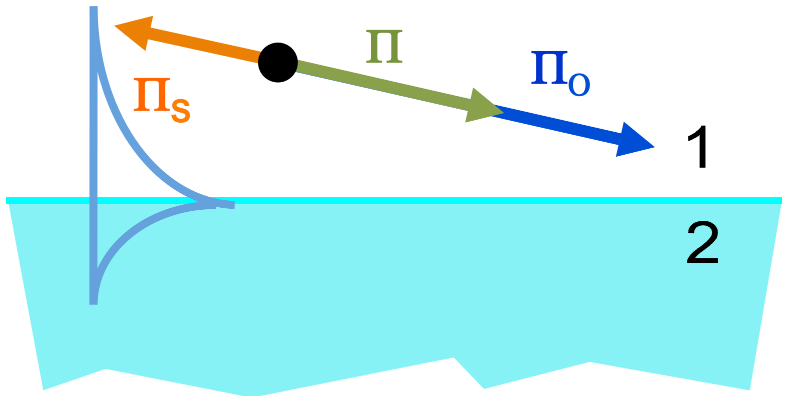
□ Poynting vector

$$\mathbf{\Pi}_S \propto e^{-2k''x}e^{-2q''z}\text{Im}[\tilde{q}\tilde{k}^\star - \tilde{k}\tilde{q}^\star][-\tilde{q}'',0,\tilde{k}'']$$

$$\mathbf{\Pi}_0 \propto e^{-2k''x}e^{-2q''z}\left(|\tilde{q}|^2 + |\tilde{k}|^2\right)[\tilde{k}',0,\tilde{q}']$$

$$\mathbf{\Pi} = \mathbf{\Pi}_O + \frac{\nabla\times\mathbf{\Phi}_{\text{SP}}}{2\omega\mu_0}$$

orbital — spin part



□ Transverse spin density

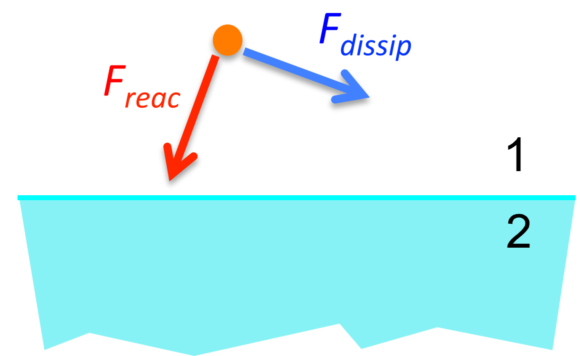
$$\mathbf{S}_{\text{SP}} = 2\frac{q'k'' - k'q''}{|\tilde{k}|^2 + |\tilde{q}|^2}\,\hat{\mathbf{y}}$$

*Canaguier-Durand & Genet, PRA **88**, 033831 (2013)*

Surface plasmon forces and torque

- SP gradient force

$$\frac{\mathbf{F}_{\text{reactive}}}{|\mathbf{E}_0|^2} = -\frac{n^2}{2}\,\mathrm{Re}[\alpha]\;[k'',0,q'']$$



- SP radiation pressure $\nabla\phi = [k',0,q']$

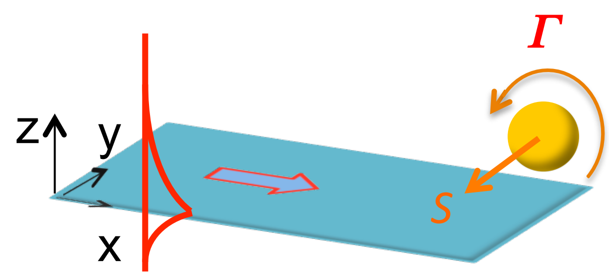
$$\frac{\mathbf{F}_{\text{dissip}}}{|\mathbf{E}_0|^2} = \frac{n^2}{2}\,\mathrm{Im}[\alpha]\;[k',0,q']$$

Confined $q' < 0$ and directed motion k'

- SP torque and transverse spin

$$\frac{\mathbf{\Gamma}}{|\mathbf{E}_0|^2} = n^2\mathrm{Im}[\alpha]\;\mathbf{S}$$

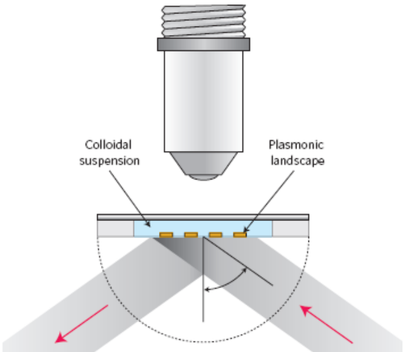
$$= -\,n^2\mathrm{Im}[\alpha]\;S\;\hat{\mathbf{y}}$$



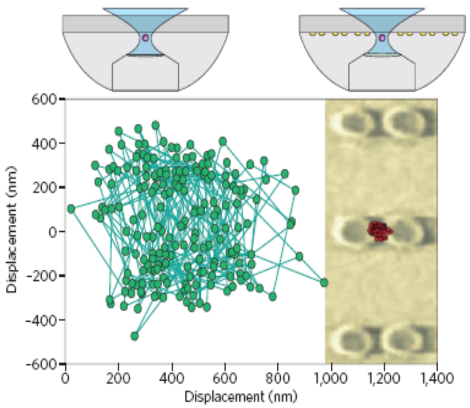
*Canaguier-Durand & Genet, PRA **89**, 033841 (2014)*

See also: Bliokh and Nori, PRA (2012)
Aiello & Banzer, arXiv 2015

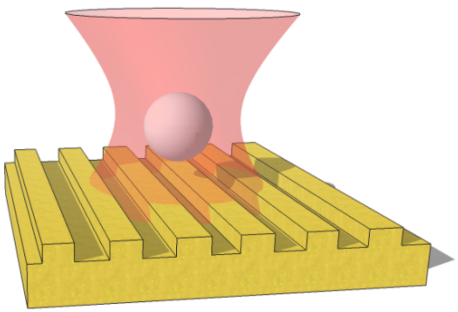
Surface plasmon-based optical tweezers



Quidant *et al.,* Nature Phys. 2007



Grigorenko *et al.* Nature Photon. 2008



Cuche *et al.* PRL 2011

« Localized » surface plasmon resonances

- *Delocalized plasmons ?*

See review Juan, Righini, Quidant Nature Photon. 2011

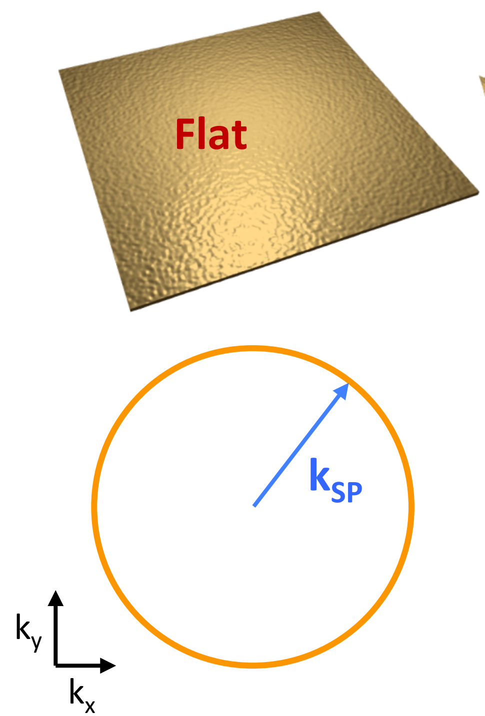
Plasmonic radiation pressure and band structures

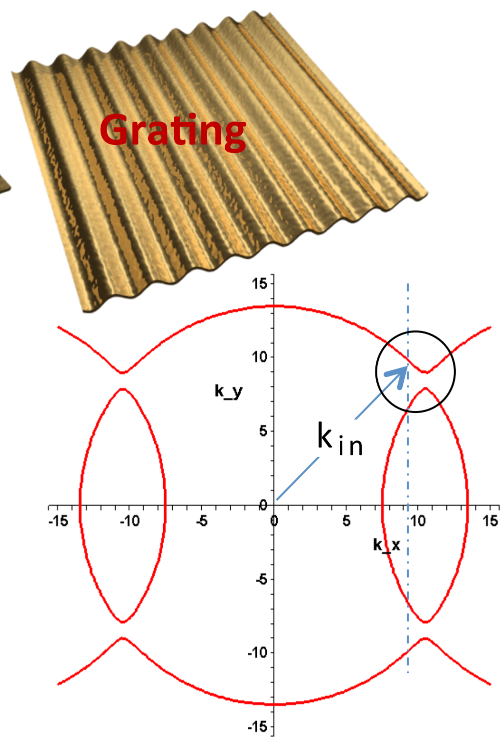
- Global energy transport

$$\langle \mathbf{\Pi}_S \rangle = \int \mathrm{d}z \mathbf{\Pi}_S = 0 \qquad \mathbf{v}_g = \frac{\langle \mathbf{\Pi}_O \rangle}{\langle W \rangle}$$

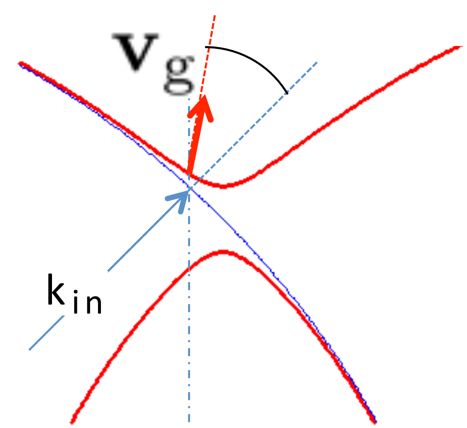
Bliokh et al., PRA (2012)

- Local anisotropy of plasmonic isofrequency surface

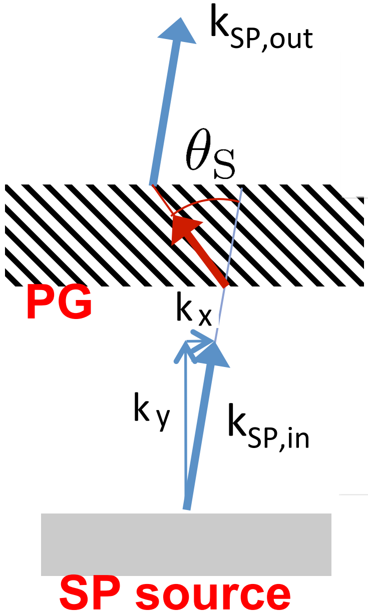


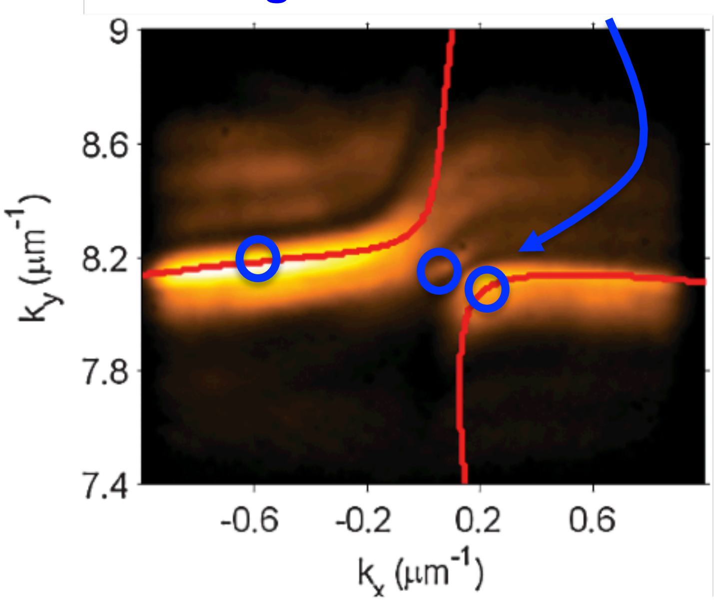


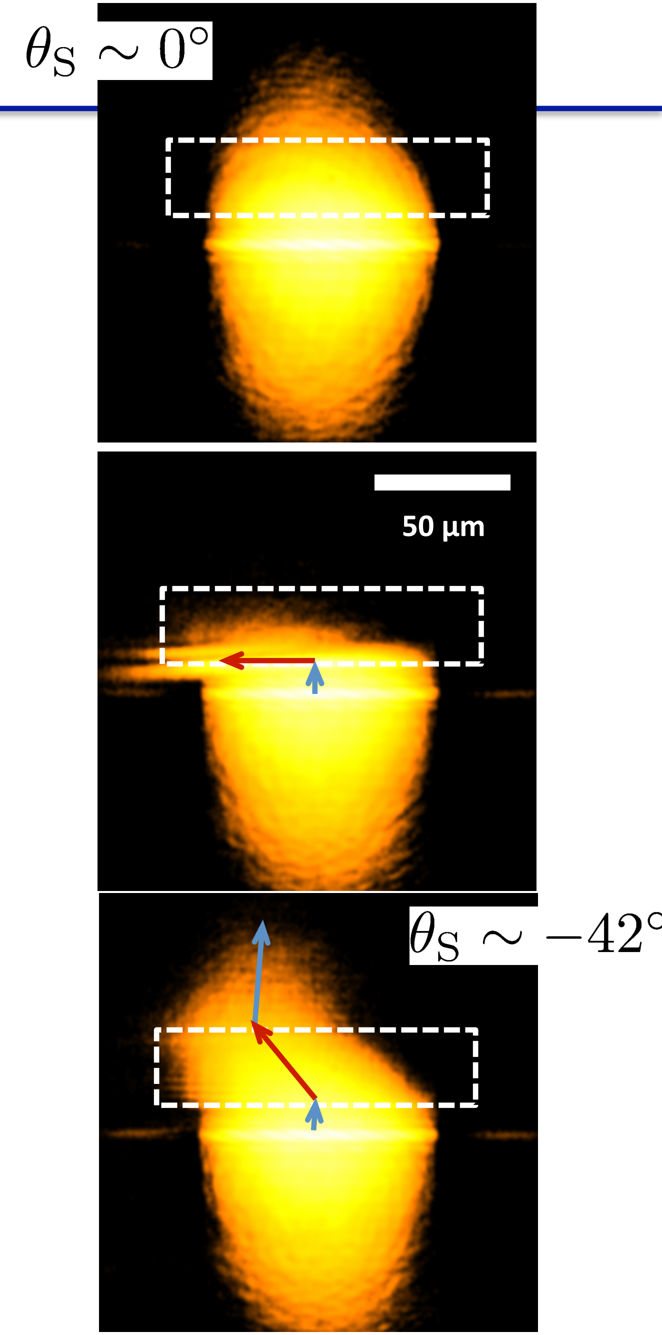
$$\mathbf{v}_\mathrm{g} = \partial_\mathbf{k} \omega(\mathbf{k})$$



Plasmonic beam steering







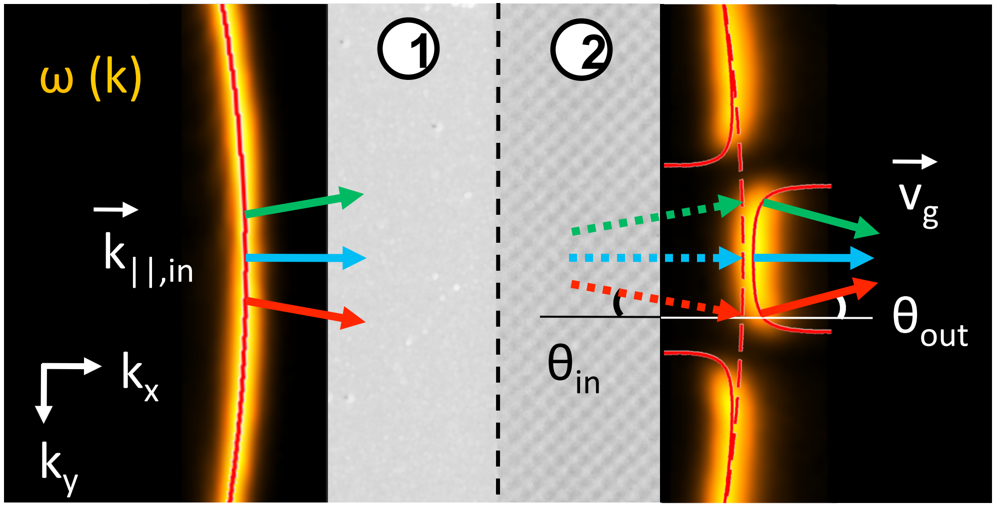
B. Stein et al., PRL ***105****, 266804 (2010)*

Dissipative plasmonic force: Bloch wave motional control

- IFS desing as a tool for controlling nanoparticle motions

① SP mode *on flat film*

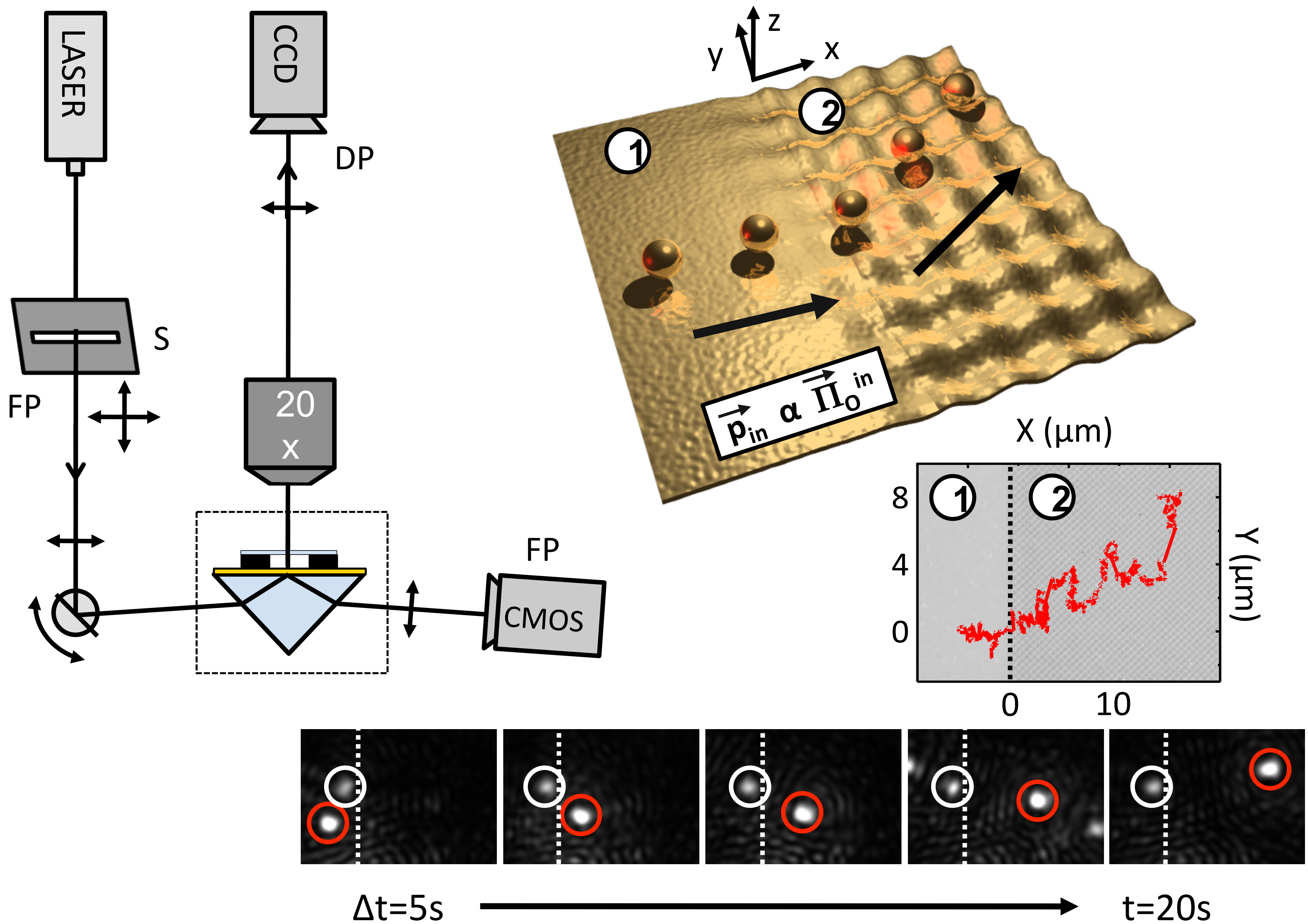
$$\mathbf{F}_{\mathrm{dissip}} \leftrightarrow \mathbf{\Pi}_O \leftrightarrow k'$$



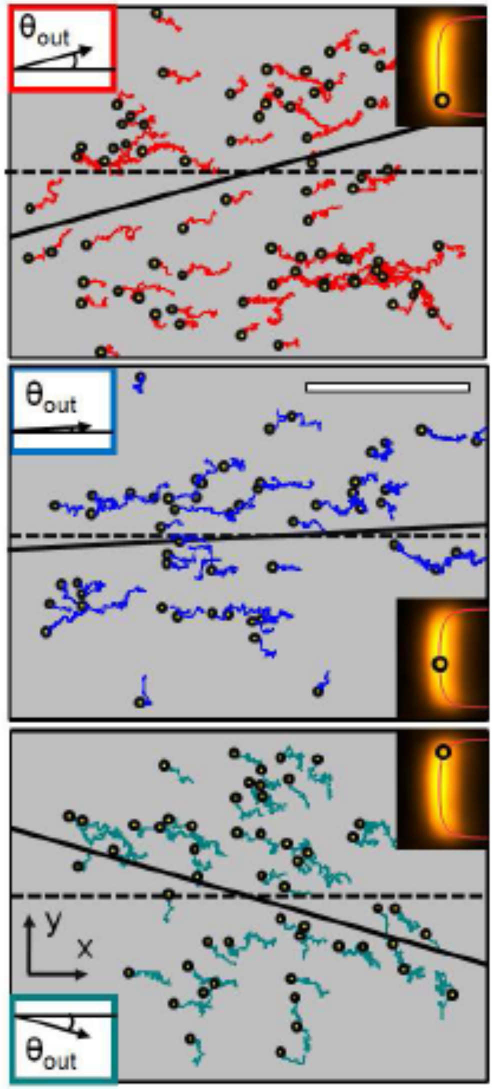
② SP *Bloch* mode

$$\mathbf{v}_{\mathrm{g}} = \nabla_{\mathbf{k}}\omega(\mathbf{k}) \leftrightarrow \langle\mathbf{\Pi}_O\rangle/\langle W\rangle \leftrightarrow \mathbf{F}_{\mathrm{dissip}}$$

Setup



Dynamical law of refraction



□ nanoparticle eq. of motion

$$\dot{\mathbf{p}} = -\frac{\gamma}{m}\mathbf{p} + \mathbf{F}_{Th} + \mathbf{F}_{\text{dissip}}$$

□ time-averaged ballistic motion

$$\overline{\mathbf{p}} = (m/\gamma)\mathbf{F}_{\text{dissip}}$$

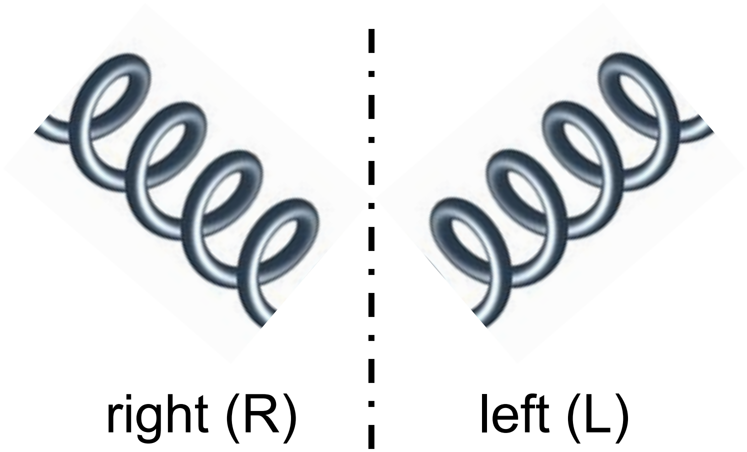
- motional evolutions determined in strict relation with the IFS
- high throughputs with high angular resolution
- mechanical analogues of *super-prism* and *negative refraction* effects

*A. Cuche et al., Nano Letters **12**, 4329 (2012)*

Chirality

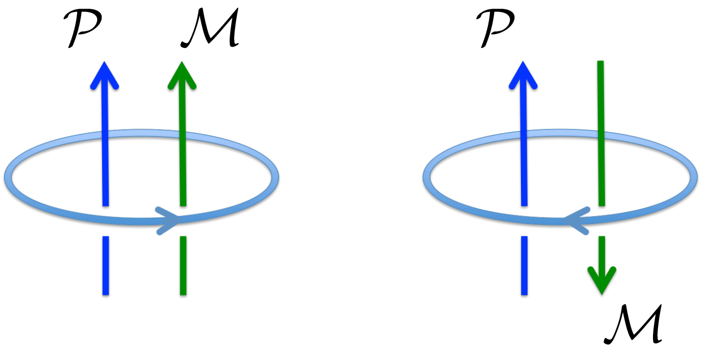
Lord Kelvin (1884) *: « I call any geometrical figure, or group of points, chiral, and say that it has chirality, if its image in a plane mirror, ideally realized, cannot be brought to coincide with itself. »*

$$[\Pi, J] \neq 0$$



Chiral dipole

- Coupled induced dipoles



$$\begin{pmatrix} \mathbf{p}_0 \\ \mathbf{m}_0 \end{pmatrix} = \begin{pmatrix} \alpha & i\chi \\ -i\chi & \beta \end{pmatrix} \begin{pmatrix} \mathbf{E}_0 \\ \mathbf{H}_0 \end{pmatrix}$$

$$[\Pi, J] \neq 0$$

Optical force and torque on a chiral dipole

Canaguier et al., NJP ***15****, 123037 (2013)*

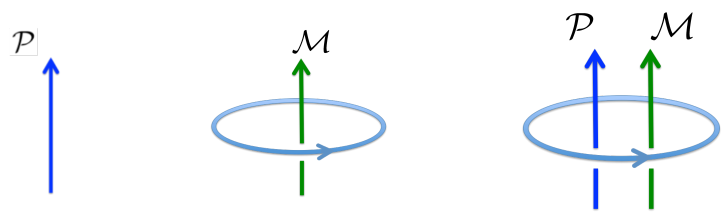
- Lorentz law (real $(\mathcal{E},\mathcal{H})$ fields)

$$\mathbf{F} = (\mathcal{P}\cdot\nabla)\mathcal{E} + (\mathcal{M}\cdot\nabla)\mathcal{H} + \mu_0\dot{\mathcal{P}}\times\mathcal{H} - \varepsilon_0\dot{\mathcal{H}}\times\mathcal{E}$$

$$\boldsymbol{\Gamma} = \mathcal{P}\times\mathcal{E} + \mathcal{M}\times\mathcal{H}$$

- Time-averaged force and torque

$$\langle\mathbf{F}\rangle_T = \frac{1}{2}\mathrm{Re}[\alpha\mathbf{f}_0] + \frac{1}{2}\mathrm{Re}[\beta\mathbf{g}_0] + \frac{1}{2}\mathrm{Re}[\chi\mathbf{h}_0]$$



$$\begin{pmatrix}\mathbf{p}_0\\ \mathbf{m}_0\end{pmatrix} = \begin{pmatrix}\alpha & i\chi\\ -i\chi & \beta\end{pmatrix}\begin{pmatrix}\mathbf{E}_0\\ \mathbf{H}_0\end{pmatrix}$$

$$\langle\boldsymbol{\Gamma}\rangle_T = \mathrm{Im}[\alpha]\boldsymbol{\Phi}_E + \mathrm{Im}[\beta]\boldsymbol{\Phi}_H + 2\mathrm{Im}[\chi]\boldsymbol{\Pi}$$

Chiral force on a chiral dipole

- Chiral part $$\langle \mathbf{F}_\chi \rangle_T = \frac{1}{2}\mathrm{Re}[\chi]\mathrm{Re}[\mathbf{h}_0] - \frac{1}{2}\mathrm{Im}[\chi]\mathrm{Im}[\mathbf{h}_0]$$

  Reactive component: $$\mathbf{F}_\chi^{r.} = \mathrm{Re}[c\chi]\frac{c}{\omega}\nabla K$$

  Dissipative component: $$\mathbf{F}_\chi^{d.} = \mathrm{Im}[c\chi]\frac{2}{c}\left(\mathbf{\Phi} - \frac{1}{2}\nabla \times \mathbf{\Pi}\right)$$

- Enantioselective forces: chiral separation
- Field chiral quantities $$K(\mathbf{r}) = \frac{\omega}{2c^2}\mathrm{Im}[\mathbf{E}_0 \cdot \mathbf{H}_0^*]$$

$$\mathbf{\Phi}(\mathbf{r}) = \frac{\omega\varepsilon_0}{4}\left(\mathrm{Im}[\mathbf{E}_0^* \times \mathbf{E}_0] + \mathrm{Im}[\mathbf{H}_0^* \times \mathbf{H}_0]\right)$$

See also: Cameron, Barnett, Yao, NJP (2014)

Optical chirality

- Free-field conservation:

Lipkin (1964)
Tang and Cohen (2010)
Bliokh and Nori (2011)
Barnett et al. NJP (2012)

Density $K(\mathbf{r},t) = \frac{\varepsilon_0\mu_0}{2}\left(\mathcal{H}\cdot\dot{\mathcal{E}} - \mathcal{E}\cdot\dot{\mathcal{H}}\right)$

Flow $\mathbf{\Phi}(\mathbf{r},t) = \frac{\varepsilon_0}{2}\mathcal{E}\times\dot{\mathcal{E}} + \frac{\mu_0}{2}\mathcal{H}\times\dot{\mathcal{H}}$

Cons. $\nabla\cdot\mathbf{\Phi} + \partial_t K = 0$

Energy and momentum

$W(\mathbf{r},t) = \frac{\varepsilon_0}{2}\mathcal{E}\cdot\mathcal{E} + \frac{\mu_0}{2}\mathcal{H}\cdot\mathcal{H}$

$\mathcal{S}(\mathbf{r},t) = \mathcal{E}\times\mathcal{H}$

$\nabla\cdot\mathcal{S} + \partial_t W = 0$

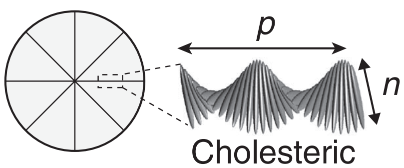
- Harmonic fields

$$K(\mathbf{r}) = \frac{\omega}{2c^2}\mathrm{Im}[\mathbf{E}_0\cdot\mathbf{H}_0^*] \qquad \mathbf{\Phi}(\mathbf{r}) = \frac{\omega\varepsilon_0}{4}\left(\mathrm{Im}[\mathbf{E}_0^*\times\mathbf{E}_0] + \mathrm{Im}[\mathbf{H}_0^*\times\mathbf{H}_0]\right)$$

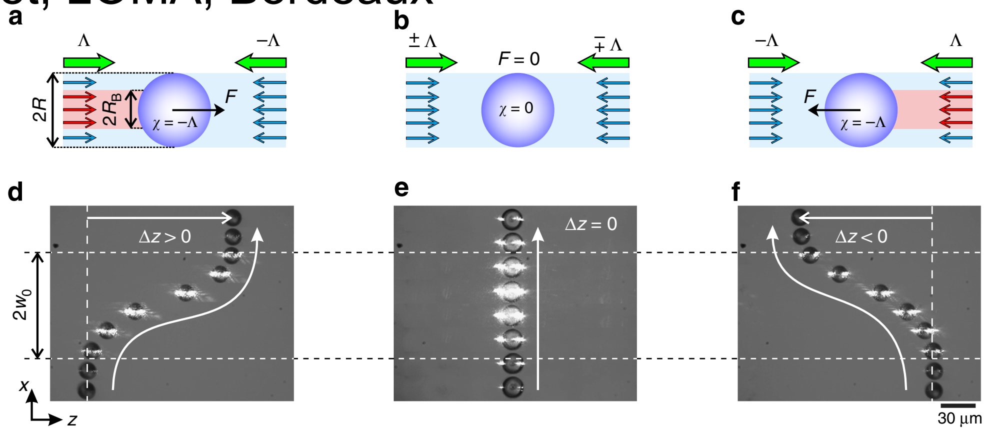
- Circularly polarized light $K = \frac{\omega I_0}{2c^2}$ $\mathbf{\Phi}_\pm = \pm\frac{\omega I_0}{2c}\hat{z}$

Chiral separation

- Tkachenko and Brasselet, LOMA, Bordeaux



radius ~ 20 microns



Nat. Commun. (2014)

- Downscaling to the nanoscale ?

$\chi/\alpha \ll 1$ ✗

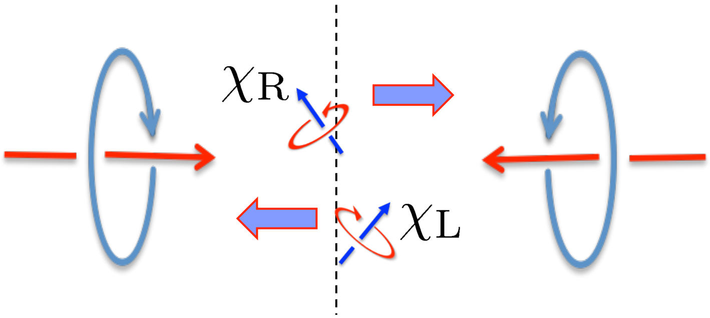
$\langle \mathbf{F}_{\alpha,\beta} \rangle$ ✗ ✓

$k_B T$ ✗ ✓

Brownian approach

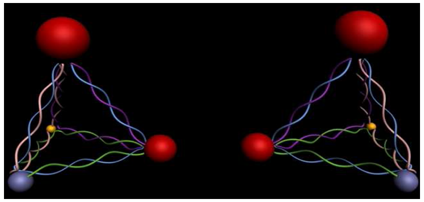
Canaguier et al., NJP **15**, *123037 (2013)*

- Cancelling $\langle \mathbf{F}^{r.}_{\alpha,\beta} \rangle$



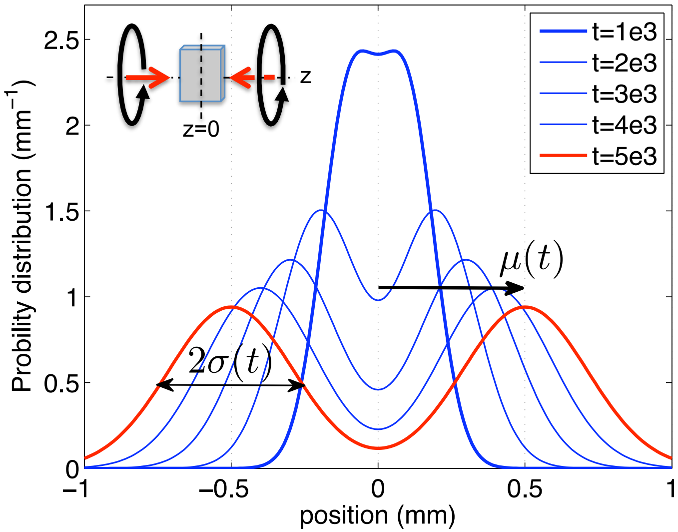
$$\langle \mathbf{F}^{d.}_{\chi} \rangle = \frac{\omega I_0}{c^2} \mathrm{Im}[c\chi]\hat{\mathbf{z}}$$

Ca. 1 mm / 1 hour
50 mW on 1 mm^2,
for *DNA-nanoparticle hybrids*
(N. Kotov, JACS 2012)



- Statistical resolution

Cuche et al., Nano Letters **12**, *4329 (2012)*



Separation $\mu(t) = (\langle F_\chi \rangle / \gamma) t$

Brownian motion causes a variance

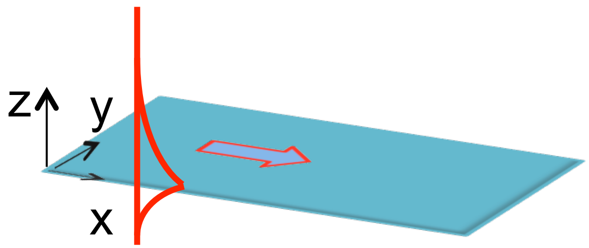
$$\sigma(t) = \sqrt{k_B/\gamma}\sqrt{t}$$

Minimal time $t_{min} \sim k_B T \gamma / \langle F_\chi \rangle^2$

Increasing optical forces: strong gradients in the near field

- Generic surface plasmon

$$\mathbf{E}_{\rm SP} = E_0[\tilde{q}, 0, -\tilde{k}]e^{ikx}e^{iqz}$$



$$K = 0$$

$$\mathbf{\Phi} = \frac{\omega I_0}{2c} e^{-2k''x-2q''z}\mathrm{Im}[\tilde{k}\tilde{q}^\star]\hat{\mathbf{y}}$$

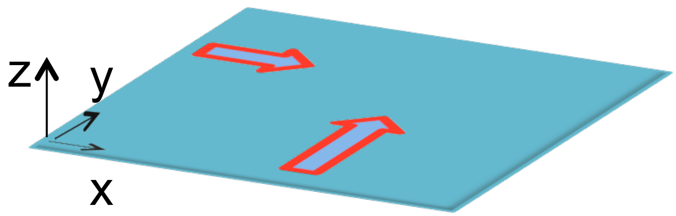
$$\mathbf{\Phi} = \frac{1}{2}\nabla \times \mathbf{\Pi}$$

No chiral force in the dipolar regime

Plasmonic optical lattice

Canaguier et al. PRA ***90****, 023842 (2014)*

- Coherent superposition $\mathbf{H}_{\mathrm{SP}} = [H_2 e^{iky}, H_1 e^{ikx}, 0] e^{iqz}$



Local phase difference $\phi(x,y) = k'(x-y)$

- Coupling terms

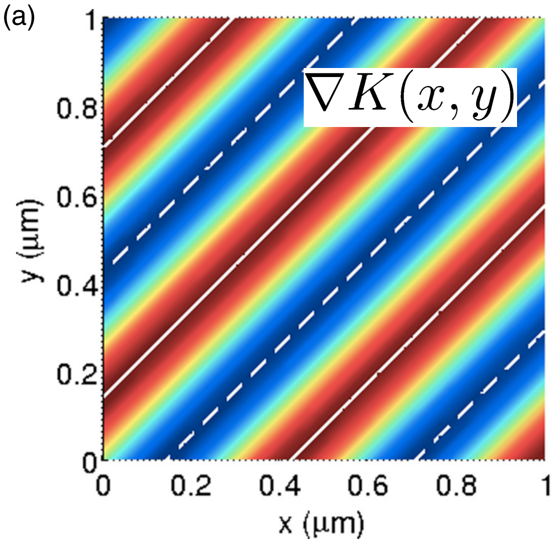
$$K(\mathbf{r}) \propto \tilde{q}' e^{-k''(x+y)} e^{-2q''z} \sin\phi \qquad \langle \mathbf{F}^{r.}_{\chi} \rangle \neq \mathbf{0}$$

$$\mathbf{\Phi}_{12} - \nabla \times \mathbf{\Pi}_{12}/2 \;\propto \tilde{q}' e^{-2k''x-2q''z} [\mathrm{Im}[\tilde{k} e^{i\phi}], \mathrm{Im}[\tilde{k}^{\star} e^{i\phi}], 2\tilde{q}' \sin\phi] \qquad \langle \mathbf{F}^{d.}_{\chi} \rangle \neq \mathbf{0}$$

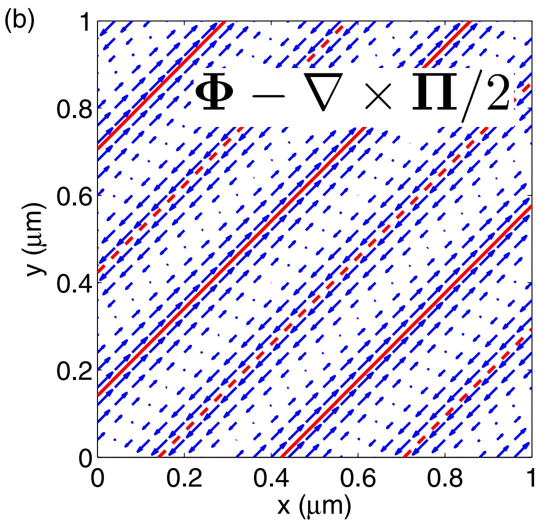
A plasmonic effect

Chiral potential energy surfaces

- Chiral force fields *(in-plane)*



Au-water interface
780 nm



Chiral potential energy

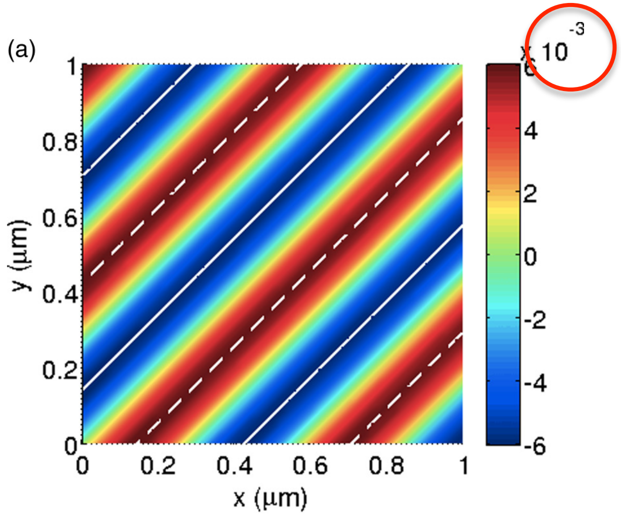
Attracts chiral dipole **towards** K_{max} / K_{min}
Pushes chiral dipole **along** K_{max} / K_{min}
} vs. $\mathrm{sgn}(\mathrm{Re}[c\chi])$

Towards near-field deracemization schemes

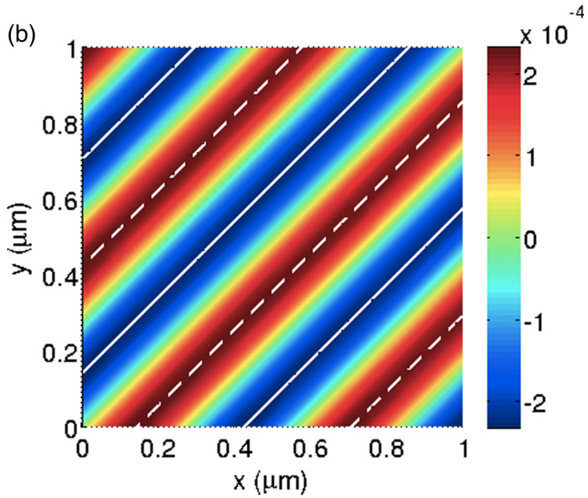
Towards chiral selective trapping

*Canaguier et al. PRA **90**, 023842 (2014)*

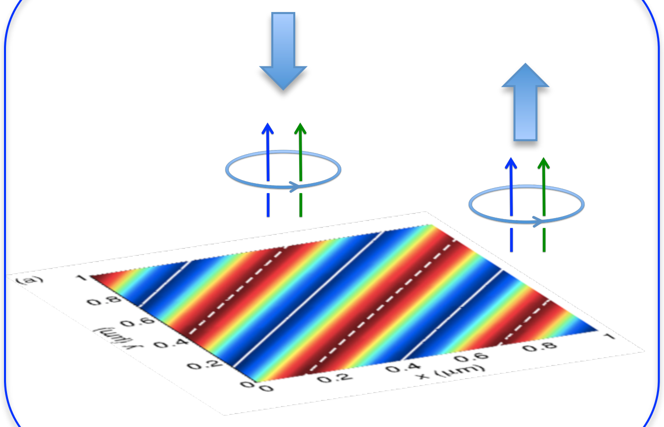
- Chiral force fields *(transverse)*



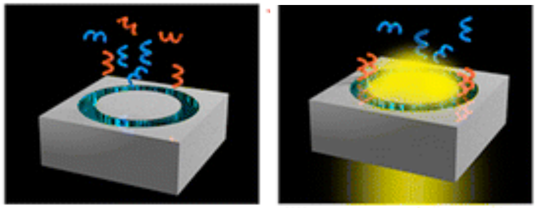
$$\nabla K \cdot \hat{\mathbf{z}}$$



$$(\mathbf{\Phi} - \nabla \times \mathbf{\Pi}/2) \cdot \hat{\mathbf{z}}$$



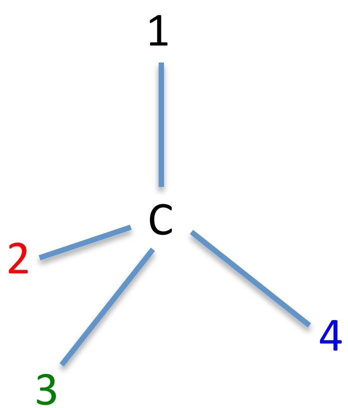
Plasmonic chiral tweezers



Dionne, ACS Photon. ASAP

Near-field enhanced chiroptical spectroscopy

Stereogenic center



□ Absorption rate of a chiral molecule

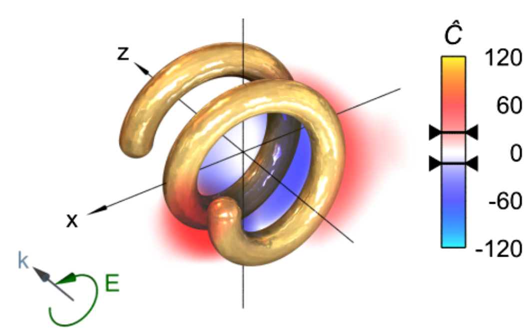
$$\epsilon_{R/L} = \langle \mathbf{E} \cdot \dot{\mathbf{p}} + \mathbf{H} \cdot \dot{\mathbf{m}} \rangle_T$$

□ Circular dichroism $\Delta\epsilon = \epsilon_L(\lambda) - \epsilon_R(\lambda) \propto \omega \cdot \mathrm{Im}[\chi]$

□ « Chiral » dichroism: Tang & Cohen, PRL 2010

$$\Delta\epsilon = \epsilon_+(\lambda) - \epsilon_-(\lambda) \propto \omega \cdot \mathrm{Im}[\chi] \cdot \mathrm{Im}[\mathbf{E} \cdot \mathbf{H}^\star]$$

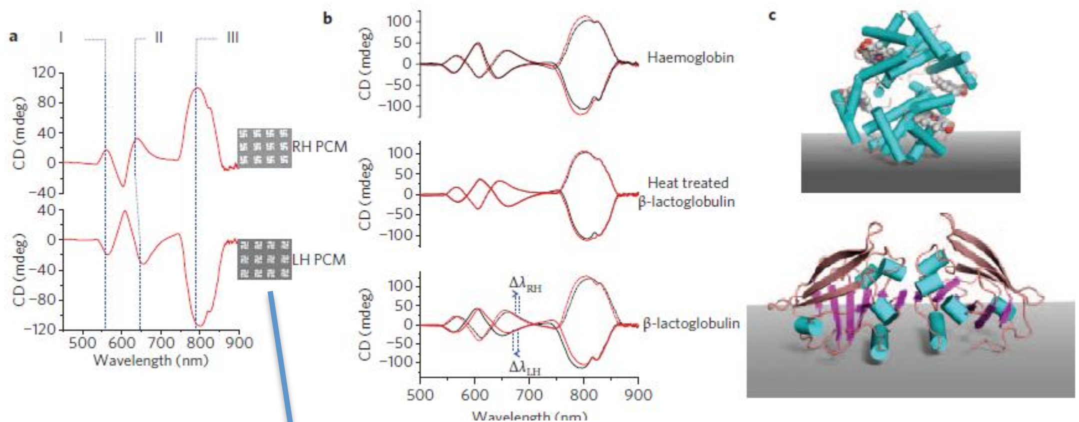
➢ « Super-chiral » fields: localized SP resonances



Hendry *et al.,* Nature Nano. (2010)
Giessen *et al.*, PR X (2012)
Barnes *et al.* PR B (2013)
Kuipers et al. Nature Phot. (2014)

Ultrasensitive detection of chiral biomolecules

E. Hendry *et al.*, Nature Nanotech. **5**, 783 (2010)



$$\Delta\epsilon_{\text{superchiral}} = 10^6 \Delta\epsilon_{\text{CPL}}$$

Near-field chirality density

Localized surface plasmons

Conclusions

- Surface plasmons and singular nano-optics
  - *New devices (ultra thin phase plates, spin splitters)*
  - *Tailoring the far field from singular near fields*
- Surface plasmon optical forces and new effects
  - *Coupling chirality of matter to chirality of light*
  - *New separation schemes based on* $K(\mathbf{r})$, $\mathbf{\Phi}(\mathbf{r})$
- Surface plasmons and enhanced chiroptical spectroscopy
  - *Inducing chiral densities on nanostructures*
  - *Matching chiral length scales light vs. molecules*