Giant Enhanced Diffusion of Nanoparticles in Optical Vortex Fields

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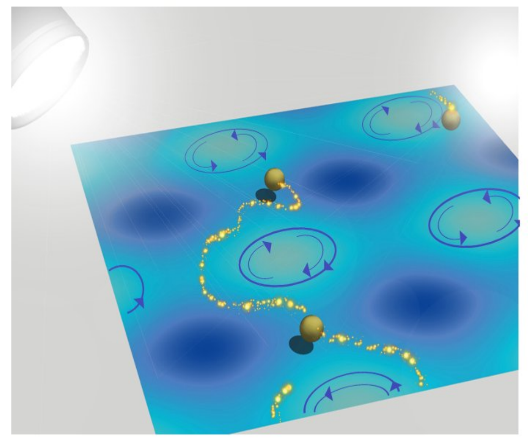
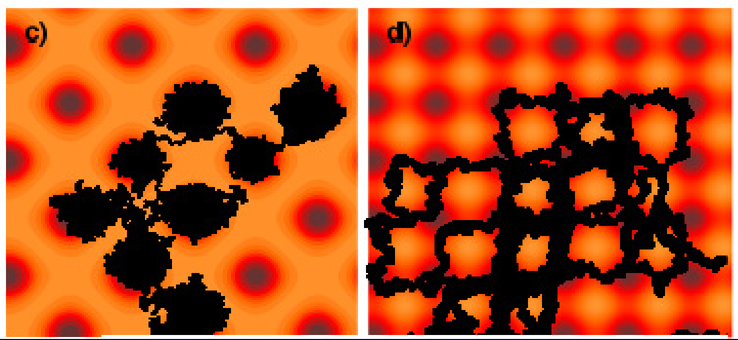
MoLE ("Moving Light & Electrons") group
(www.uam.es/mole)

[Universidad Autónoma de Madrid]





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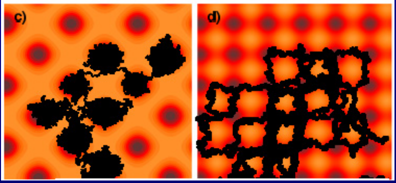
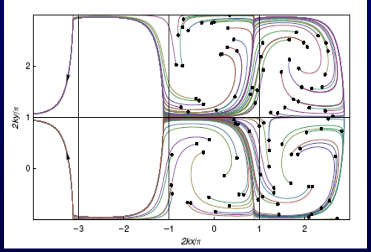
NanoLett. **9**, 3527 (2009)

- Introduction

  Optical Forces on small particles.



- Giant enhanced diffusion of gold nanoparticles on optical vortex lattices



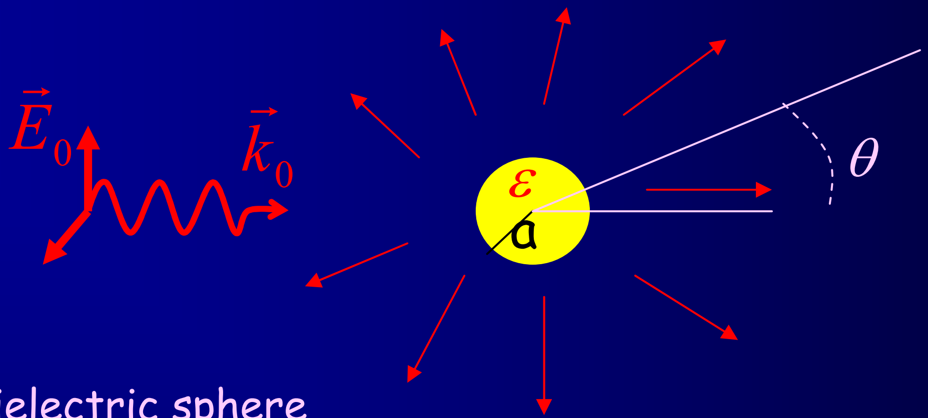
- Deterministic Ratchet from Stationary Light vortex Fields.

Scattering from a small particle

- Electric dipole

- Polarizability

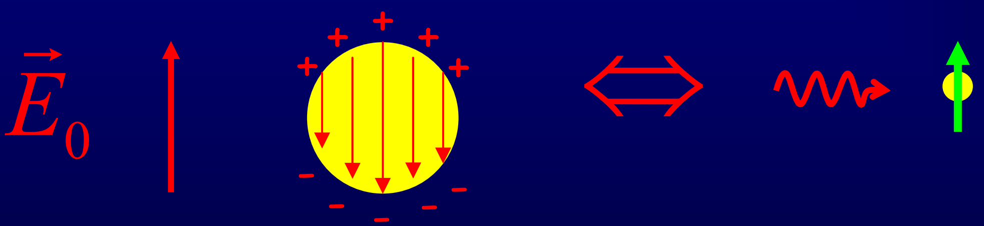
Scattering by a small particle



Small dielectric sphere
(Classical Electrodynamics, Jackson)

$\lambda >> a$

$$\vec{p} = \varepsilon_0 \varepsilon_h \alpha \vec{E}_0$$



Within the electrostatic approximation, α is **real**.

$$\alpha_0 = 4\pi a^3 \left(\frac{\varepsilon - \varepsilon_h}{\varepsilon + 2\varepsilon_h} \right)$$

Microseres

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$$\vec{p} = \varepsilon_0 \varepsilon_h \alpha \vec{E}_0$$

$$\alpha = \frac{\alpha_0}{1 - i\dfrac{k^3}{6\pi}\alpha_0}$$

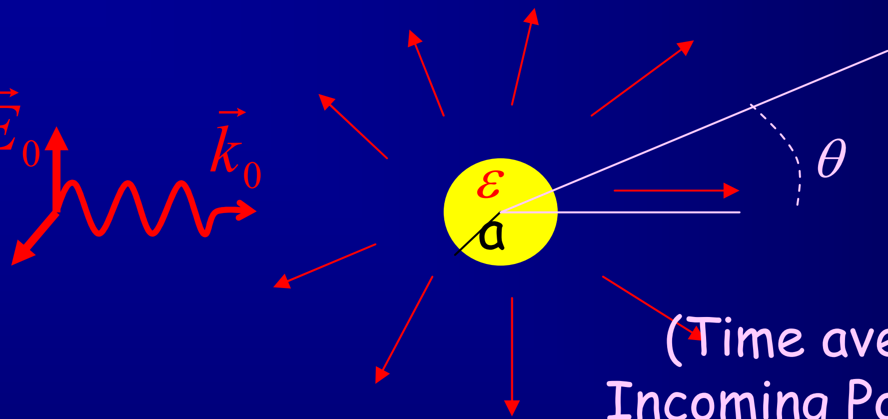
$$k\,\mathrm{Im}\{\alpha\} = \sigma$$

$$\mathrm{Re}\{\alpha\} \approx \alpha_0$$

$$\mathrm{Im}\left\{\frac{1}{\alpha}\right\} = -\frac{k^3}{6\pi} = -k^2\,\mathrm{Im}\{G_{ii}(0,0)\}$$

$$\lambda >> a$$

Let us consider plane wave incidence



(Time averaged)
Incoming Power/Area
<S>

$$\frac{1}{2}\Re\left\{\mathbf{E}\times\mathbf{H}^*\right\}$$

Scattering Cross Section =
(Scattered Power)/(Inc. Power/area)

$$\Leftrightarrow \sigma_{scat} = k^3|\alpha|^2\,\mathrm{Im}\{G_{ii}(0,0)\} = \frac{k^4}{6\pi}|\alpha|^2$$

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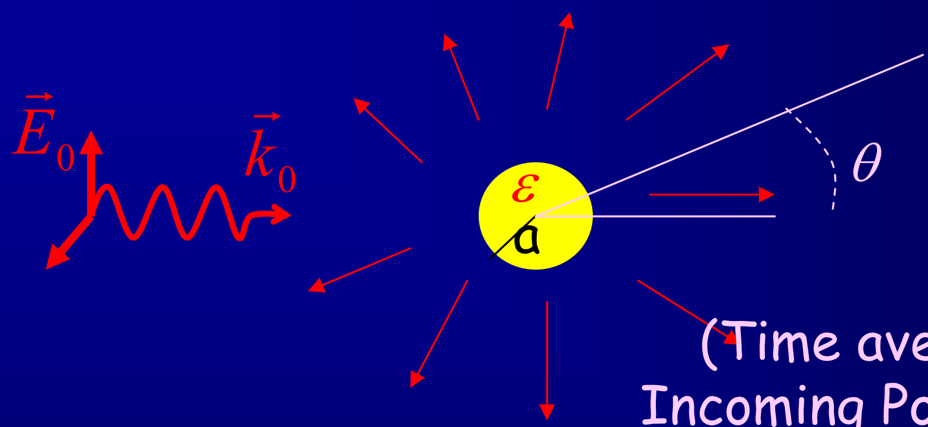
Let us consider plane wave incidence



(Time averaged)
Incoming Power/Area
<S>

$$\frac{1}{2}\Re\{\mathbf{E}\times\mathbf{H}^*\}$$

Extinction Cross Section =
(Power removed from the beam)/(Inc. Power/area)

$$\sigma_{ext} = k\,\mathrm{Im}\{\alpha\}$$

Optical Theorem

$$\sigma = k\,\mathrm{Im}\{\alpha\}$$ Extinction Cross Section

In absence of absorption:
Transmitted + Reflected Power = Incoming Power

$$\mathrm{Im}\{\alpha\} = k^2|\alpha|^2\,\mathrm{Im}\{G_{ii}\}$$

$$\mathrm{Im}\left\{\frac{1}{\alpha}\right\} = -k^2\,\mathrm{Im}\{G_{ii}(0,0)\}$$

$$\Leftrightarrow \sigma = k^3|\alpha|^2\,\mathrm{Im}\{G_{ii}(0,0)\} = \frac{k^4}{6\pi}|\alpha|^2$$

If α is real:
Power is not conserved

Optical Forces

For small particles

$$\vec{F} = \frac{d}{dt}\vec{P}_{mec} = \int_V \left(\rho\vec{E} + \vec{J}\times\vec{B}\right)dV$$

$$\left\langle \vec{F} \right\rangle_t = \frac{1}{2}\mathrm{Re}\left\{ \sum_i p_i \vec{\nabla} E_i^* \right\} =$$

$$< F >= \mathrm{Re}\{\alpha\}\left\{ \nabla \frac{1}{4}\left|E\right|^2 \right\} + \sigma \frac{1}{2}\mathrm{Re}\left\{ \frac{1}{c} E \times H^* \right\}$$

$$+ \sigma \frac{1}{2}\mathrm{Re}\left\{ i \frac{\varepsilon_0}{k_0}\left(E \cdot \nabla\right)E^* \right\}$$

????

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Optical Forces
(Maxwell eqs. + Lorentz Force)

$$\left\langle \vec{F} \right\rangle_t = \frac{1}{2}\mathrm{Re}\left\{ \sum_i p_i \vec{\nabla} E_i^* \right\} =$$

(A. Ashkin, PRL (70))

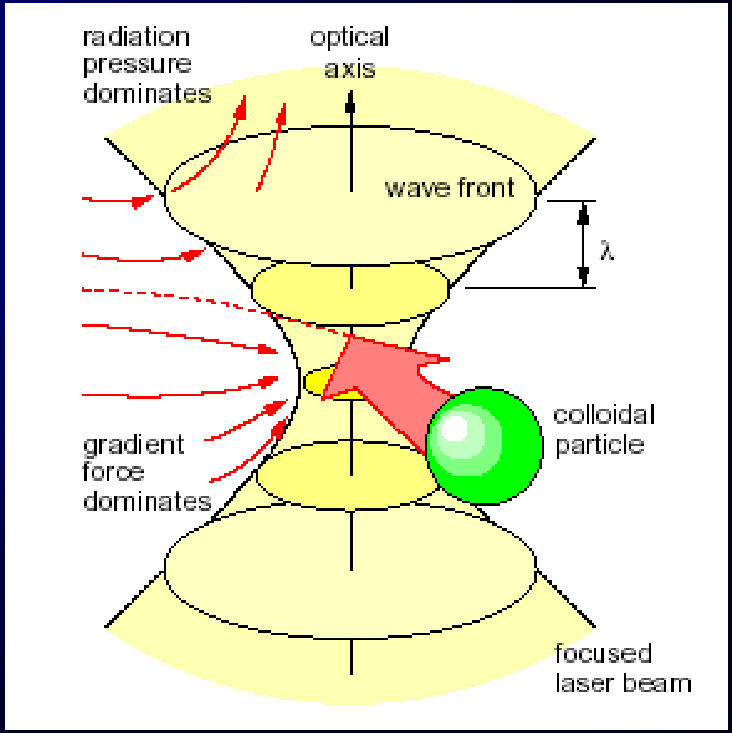
Optical Tweezers

$$\frac{1}{4}\alpha_0 \vec{\nabla}\left|\vec{E}_0\right|^2$$ Polarization

$$+$$

$$\frac{1}{c}\left\langle \vec{S} \right\rangle \sigma$$ Radiation Pressure

(J.P. Gordon, PRA (73))



D.G. Grier, Nature (03)

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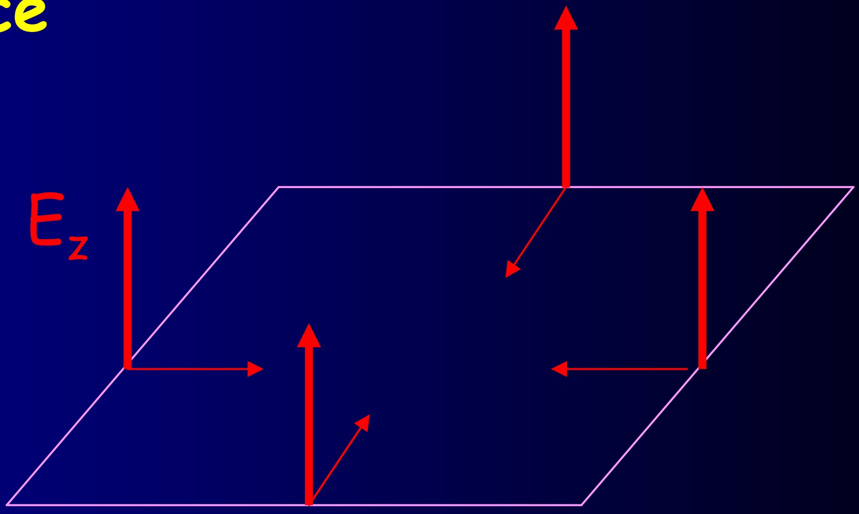
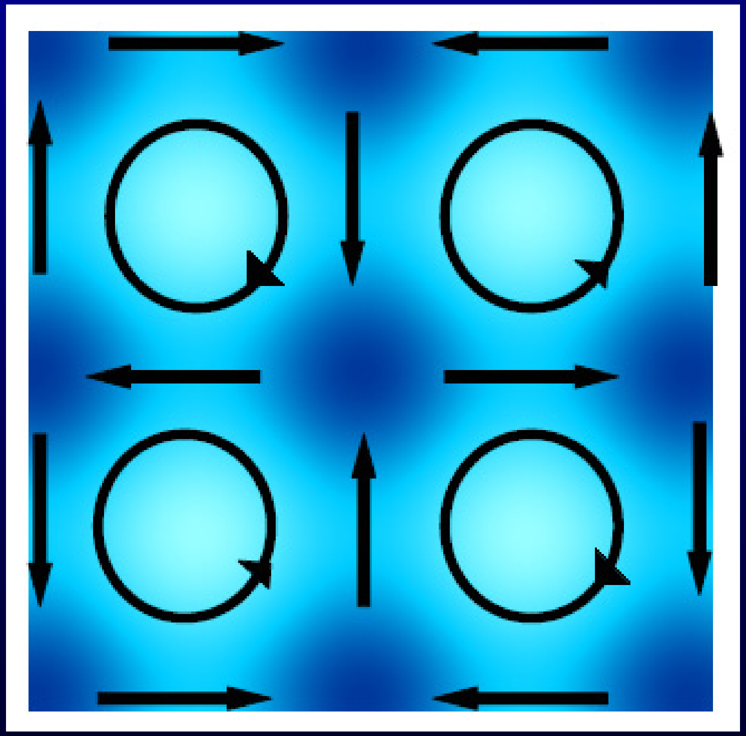
Optical vortex lattice

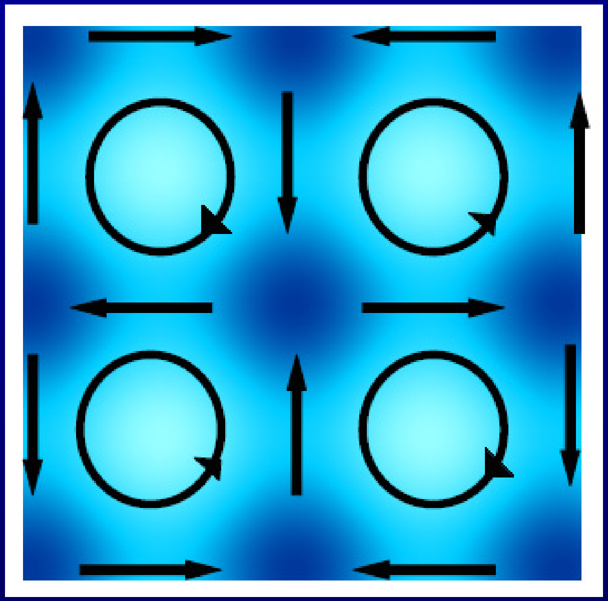


A. Hemmerich and T.W. Hänsch, Phys. Rev. Lett. 68, 1492 (**1992**)



$$E_z(x, y; \omega) = \frac{2iE_0}{\sqrt{2}}\left(\sin k_0 x + e^{i\phi} \sin k_0 y\right)$$

Optical vortex lattice



$$\mathbf{F}^{(s)} = \Re\{\alpha\}\frac{1}{2}\nabla|E_z|^2 + \sigma\left\{\frac{1}{c}\langle S\rangle^{(s)}\right\}$$

$$\frac{1}{c}\langle S\rangle^{(s)} = c\nabla \times \langle L_O\rangle$$

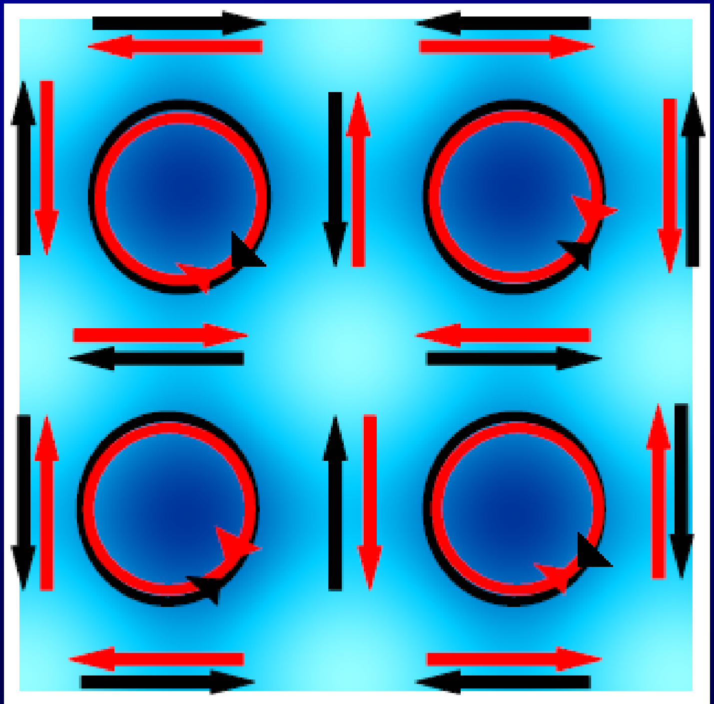
$$\langle L_O\rangle \equiv \frac{\epsilon_0}{\omega}|E_0|^2 \sin\phi\cos k_0x\cos k_0y\ \mathbf{u}_z$$

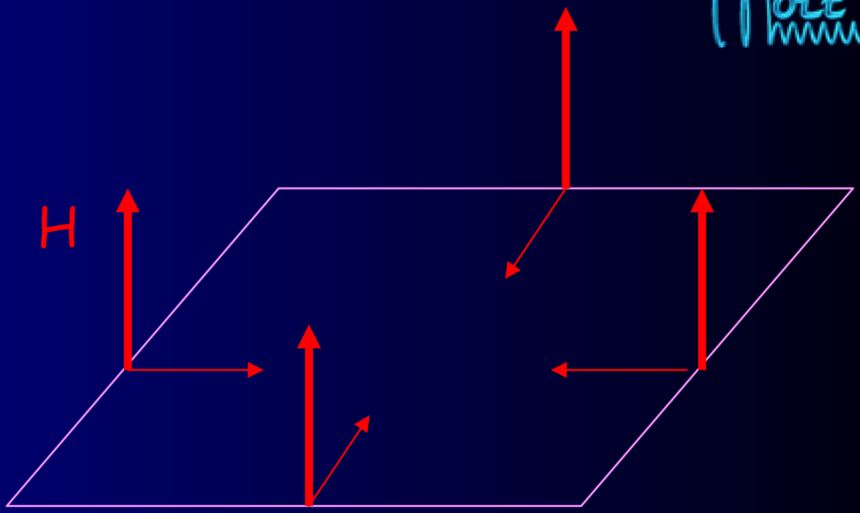
L_O => Orbital Angular Momentum

$$\frac{|E_z|^2}{|E_0|^2} = \sin^2 \mathrm{k}_0 x + \sin^2 \mathrm{k}_0 y + 2\cos\phi\sin \mathrm{k}_0 x \sin \mathrm{k}_0 y$$

The curl force can be associated to the orbital angular momentum, L_O arising as a consequence of the rotation of the Poynting vector around the field nodes.

Optical lattice

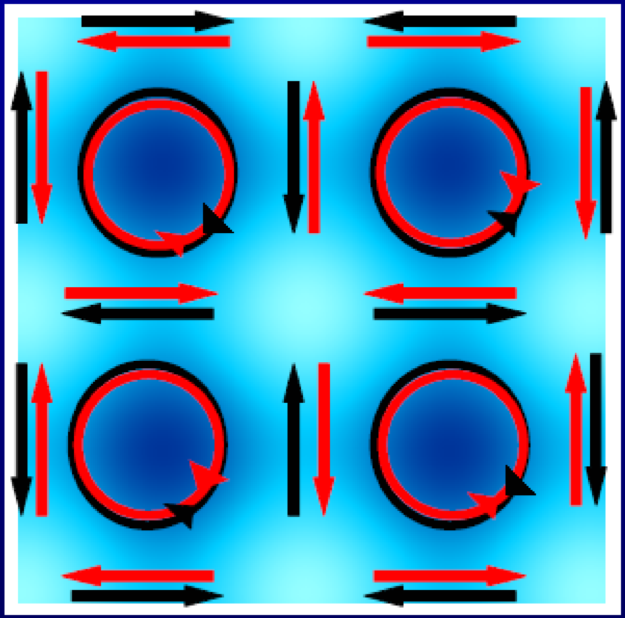




A. Hemmerich and T.W. Hänsch,
Phys. Rev. Lett. 70, 410 (**1993**)

$$H_z(x,y;\omega) = -\epsilon_0 c \frac{2iE_0}{\sqrt{2}} \left(\sin k_0 x + e^{i\phi} \sin k_0 y\right)$$

Optical lattice



$$H_z(x,y;\omega) = -\epsilon_0 c \frac{2iE_0}{\sqrt{2}} \left(\sin k_0 x + e^{i\phi} \sin k_0 y\right)$$

$$E_x = \frac{2E_0}{\sqrt{2}} e^{i\phi} \cos k_0 y$$

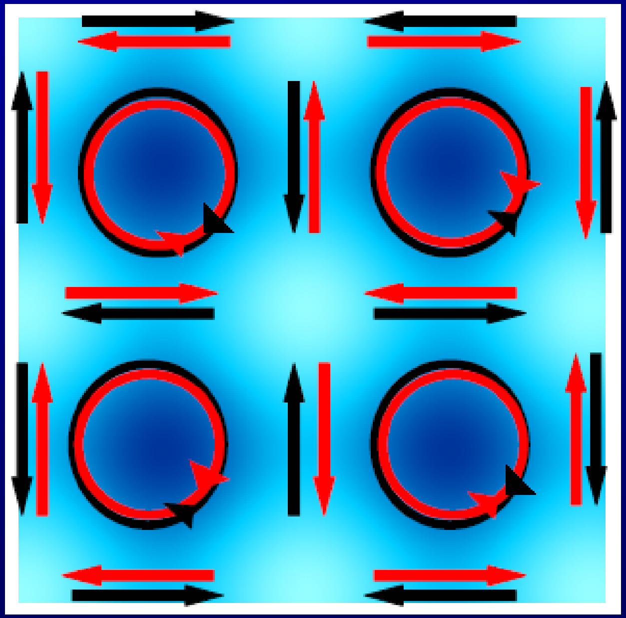
$$E_y = -\frac{2E_0}{\sqrt{2}} \cos k_0 x.$$

$$\boldsymbol{F}^{(p)} = \Re\{\alpha\}\frac{1}{2}|E_0|^2 \boldsymbol{\nabla} \left(\cos^2 k_0 x + \cos^2 k_0 y\right).$$

Does not depend on the Phase Shift !

CONSERVATIVE !

Optical lattice



$$\boldsymbol{F}^{(p)} = \Re\{\alpha\}\frac{1}{2}|E_0|^2\boldsymbol{\nabla}\left(\cos^2 k_0 x + \cos^2 k_0 y\right).$$

Poynting vector?

$$\frac{1}{c}\langle \boldsymbol{S}\rangle^{(s)} = c\boldsymbol{\nabla}\times\langle \boldsymbol{L}_O\rangle$$

$$\langle \boldsymbol{L}_O\rangle \equiv \frac{\epsilon_0}{\omega}|E_0|^2 \sin\phi \cos k_0 x \cos k_0 y \ \mathbf{u}_z$$

Microseres

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Optical Forces
(Maxwell eqs. + Lorentz Force)

$$\left\langle \vec{F} \right\rangle_t = \frac{1}{2}\mathrm{Re}\left\{ \sum_i p_i \vec{\nabla} E_i^* \right\} =$$

Polarization

$$\langle F \rangle = \frac{1}{4}\Re\{\alpha\}\nabla|\mathbf{E}|^2 + \sigma\frac{1}{2}\Re\left\{\frac{1}{c}\mathbf{E}\times\mathbf{H}^*\right\} + \sigma\frac{1}{2}\Re\left\{i\frac{\epsilon_0}{k_0}(\mathbf{E}^*.\nabla)\mathbf{E}\right\}$$

???

Radiation Pressure

$$\frac{1}{c}\left\langle \vec{S} \right\rangle \sigma$$

Microseres

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Optical Forces

(Maxwell eqs. + Lorentz Force)

$$\left\langle \vec{F} \right\rangle_t = \frac{1}{2} \mathrm{Re}\left\{ \sum_i p_i \vec{\nabla} E_i^* \right\} =$$

Polarization

$$\langle \boldsymbol{F} \rangle = \frac{1}{4} \Re\{\alpha\} \boldsymbol{\nabla} |\mathbf{E}|^2 + \sigma \frac{1}{2} \Re\left\{ \frac{1}{c} \mathbf{E} \times \mathbf{H}^* \right\} + \sigma \frac{1}{2} \Re\left\{ i \frac{\epsilon_0}{k_0} (\mathbf{E}^* . \boldsymbol{\nabla}) \mathbf{E} \right\}$$

$$\sigma \frac{1}{2} \Re\left\{ i \frac{\epsilon_0}{k_0} (\mathbf{E}^* . \boldsymbol{\nabla}) \mathbf{E} \right\} = \sigma c \boldsymbol{\nabla} \times \left(\frac{\epsilon_0}{4\omega i} (\mathbf{E} \times \mathbf{E}^*) \right)$$

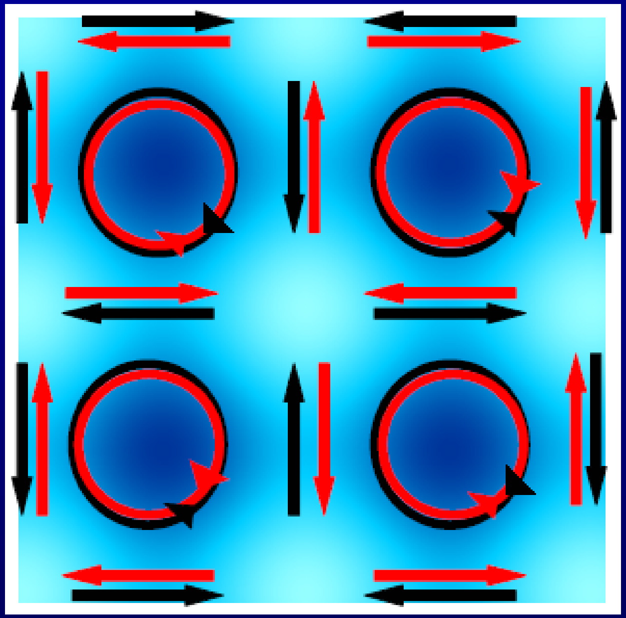
Radiation Pressure

$$\frac{1}{c} \left\langle \vec{S} \right\rangle \sigma$$

$$\langle \boldsymbol{L}_S \rangle = \frac{\epsilon_0}{4\omega i} \{\mathbf{E} \times \mathbf{E}^*\}$$

Time averaged spin density!!

Optical lattice



$$F^{(p)} = \Re\{\alpha\}\frac{1}{2}|E_0|^2\nabla\left(\cos^2 k_0x + \cos^2 k_0y\right).$$

Poynting vector?

$$\frac{1}{c}\langle S\rangle^{(s)} = c\nabla\times\langle L_O\rangle$$

$$\langle L_O\rangle \equiv \frac{\epsilon_0}{\omega}|E_0|^2\sin\phi\cos k_0x\cos k_0y\ \mathbf{u}_z$$

$\langle L_S\rangle$ = - $\langle L_O\rangle \equiv \frac{\epsilon_0}{\omega}|E_0|^2\sin\phi\cos k_0x\cos k_0y\ \mathbf{u}_z$

Gradient force

Scattering force

$$\langle \boldsymbol{F} \rangle = \Re\{\alpha\} \left\{ \boldsymbol{\nabla} \frac{1}{2} \langle |\mathrm{E}|^2 \rangle \right\} + \sigma \left\{ \frac{1}{c} \langle \boldsymbol{S} \rangle \right\} + \sigma \left\{ c \boldsymbol{\nabla} \times \langle \boldsymbol{L}_S \rangle \right\}.$$

The scattering force (proportional to the total cross section) can be written as the sum of two contributions:

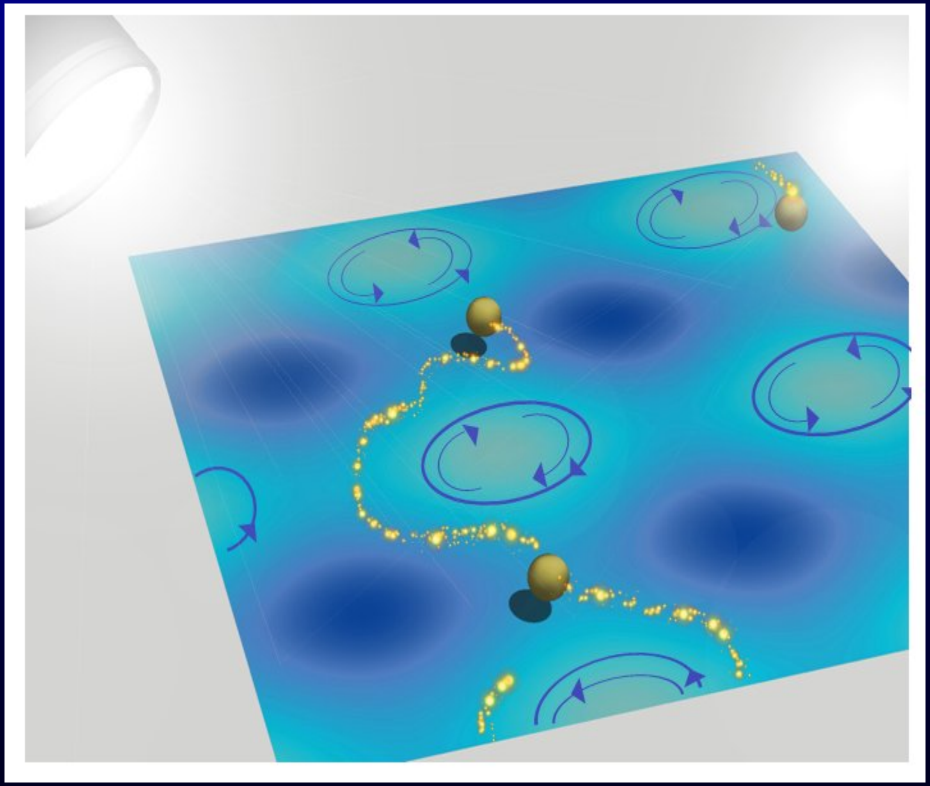
-the traditional radiation pressure term, proportional to the Poynting vector,
-a curl force associated to the non-uniform distribution of the spin density of the light field.

When the light is linearly polarized the curl term is identically zero.

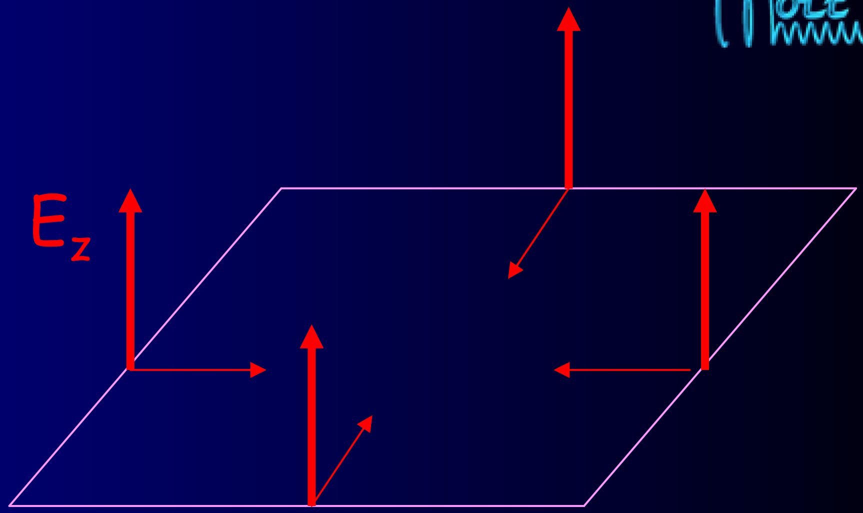
(S. Albaladejo et al., PRL (2009))

Giant enhanced diffusion of gold nanoparticles on vortex lattices

(S. Albaladejo et al., Nano Letters (2009))



Optical vortex lattice



A. Hemmerich and T.W. Hänsch,
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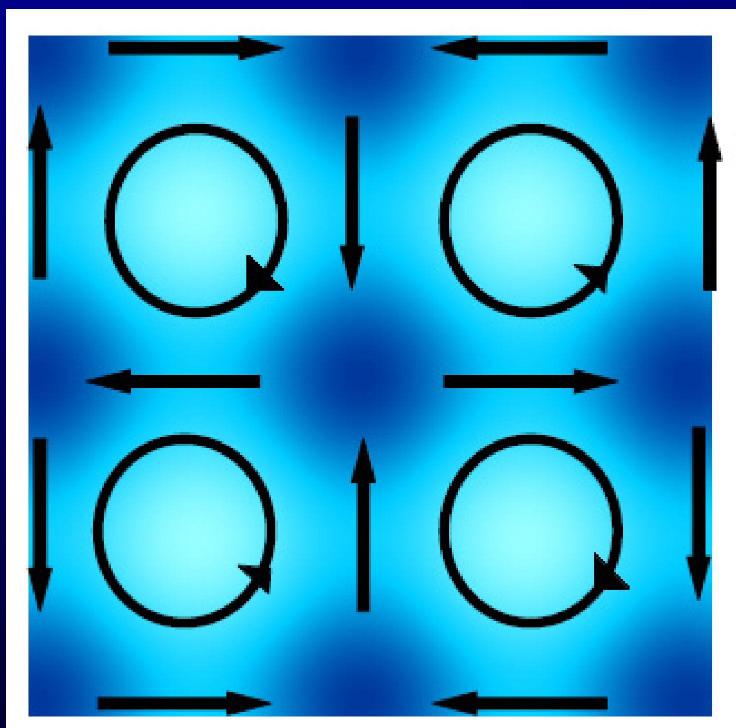
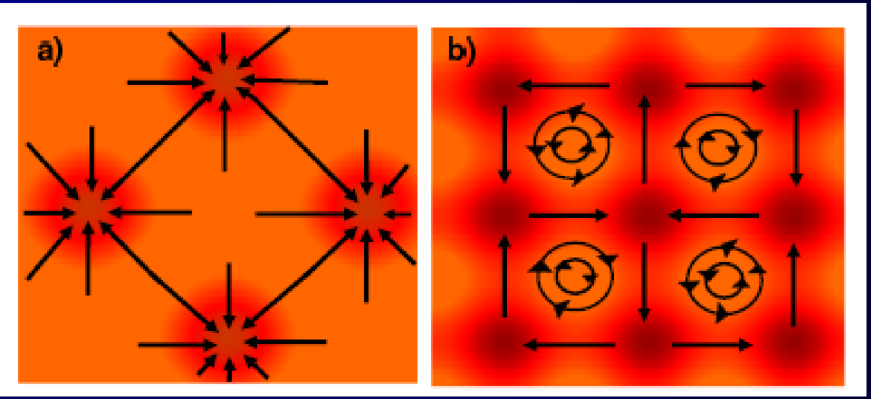
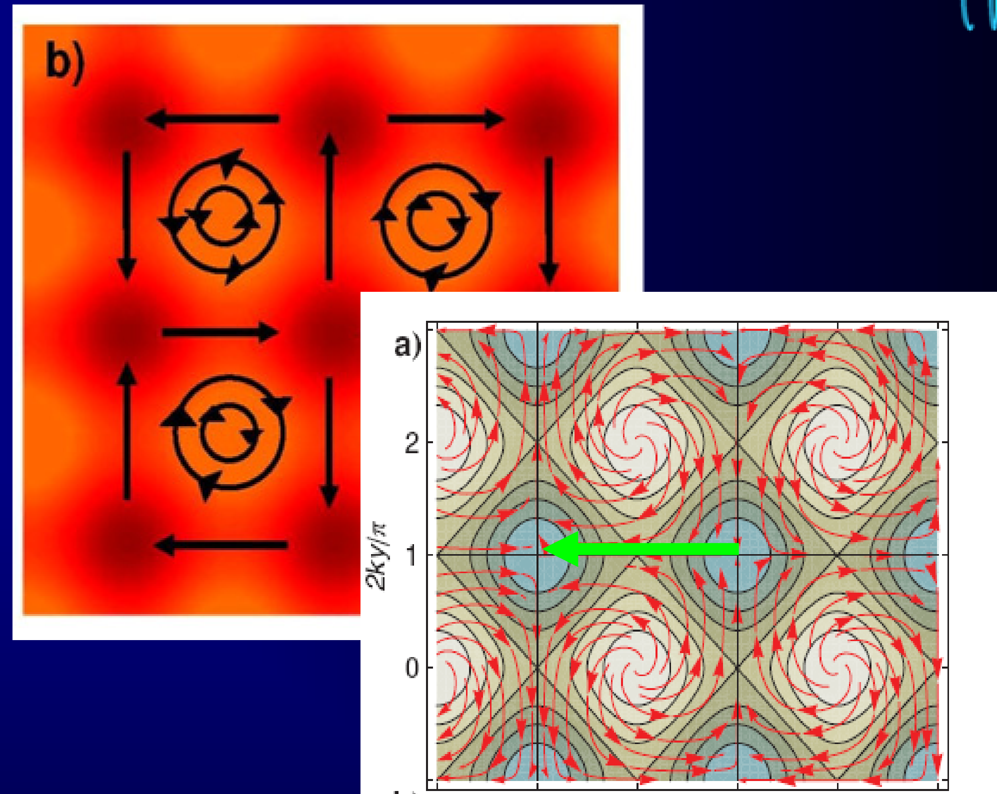


$$(n/c)P \equiv \varepsilon_0 \varepsilon |E_0|^2/2.$$

$$F_{\phi-0} = 2\alpha'(n/c)P\nabla(\sin kx + \sin ky)^2$$



$$F_{\phi=\pi/2} = 2\alpha'(n/c)P\nabla(\sin^2 kx + \sin^2 ky) + 2\alpha''(n/c)P\nabla \times \{2 \cos kx \cos ky u_z\}$$



$$4(n/c)P(\alpha' \sin kx - \alpha'')\cos kx.$$

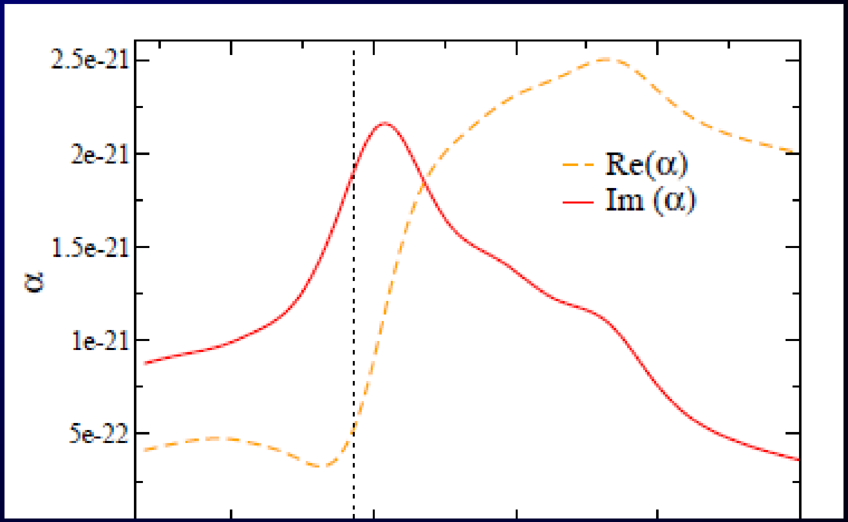
$$\alpha'' > \alpha',$$

NO Stable positions in the system !!!

Gold nanoparticles

$$\alpha_0 = 4\pi\varepsilon_0 a^3 \left(\frac{\varepsilon(\omega)-1}{\varepsilon(\omega)+2} \right)$$

$$\alpha = \frac{\alpha_0}{1 - i\dfrac{k^3}{6\pi}\alpha_0}$$



530 nm

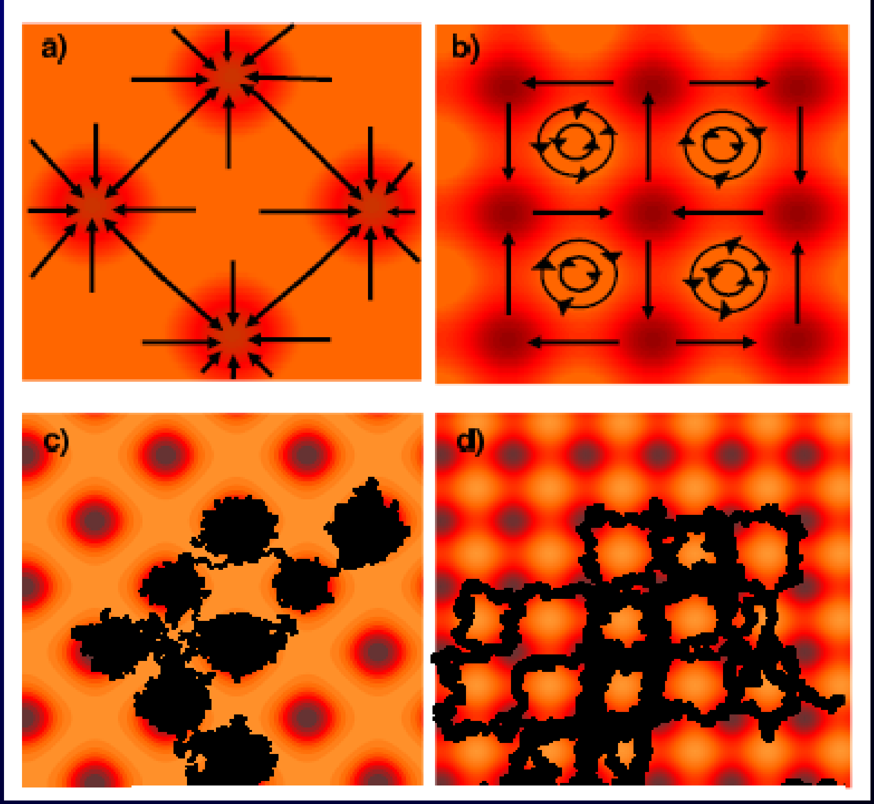
$$m\frac{d^2r}{dt^2} = F(r) - \gamma\frac{dr}{dt} + \xi(t)$$

$$\gamma = 6\pi a\eta$$

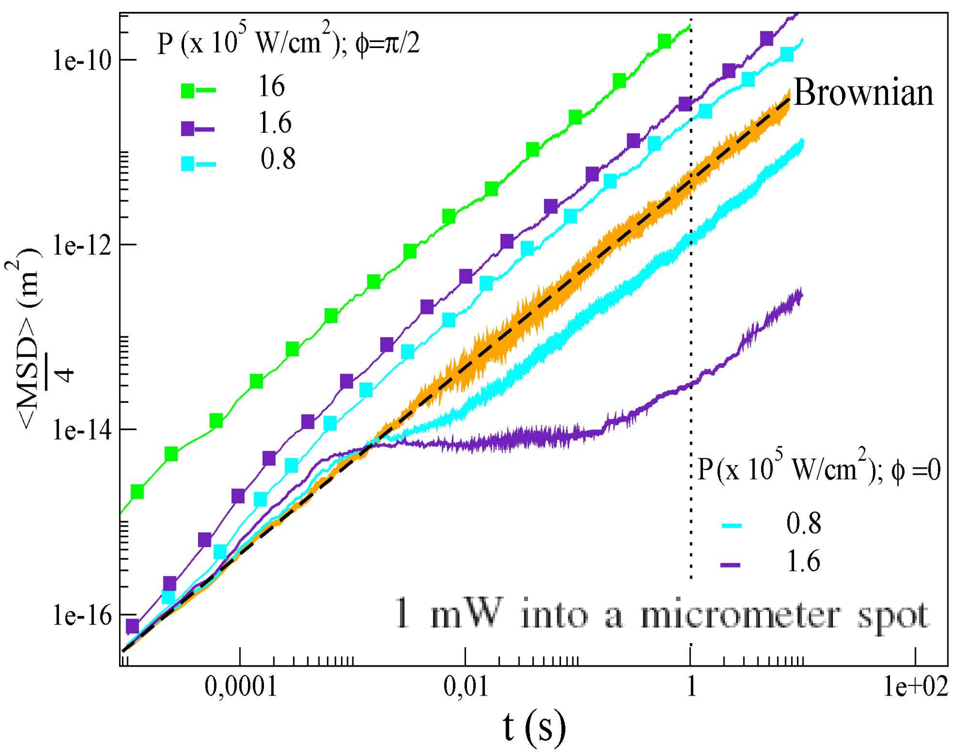
$$T = 298\ \mathrm{K},\ \eta = 0.89 \times 10^{-3}\ \mathrm{kg\ m^{-1}\ s^{-1}}$$

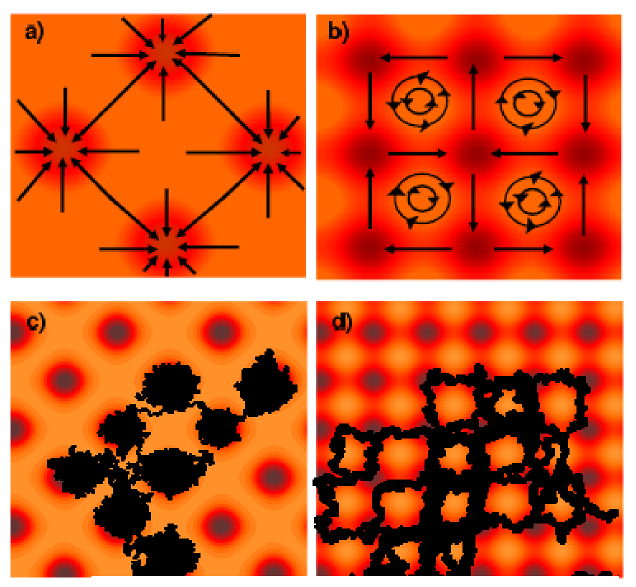
$$\langle \xi_i(t)\xi_j(t')\rangle = 2\gamma k_B T \delta_{ij}\delta(t - t').$$

Langevin dynamics



50 nm gold nanoparticles in water





$$\langle |x(t) - x(0)|^2 + |y(t) - y(0)|^2 \rangle = \langle r^2 \rangle = 4Dt$$

(S. Albaladejo et al. NanoLett. 2009)

$$D_0 = k_B T/\gamma \approx 4.9 \times 10^{-12}\ \mathrm{m^2/s}.$$

Microseres

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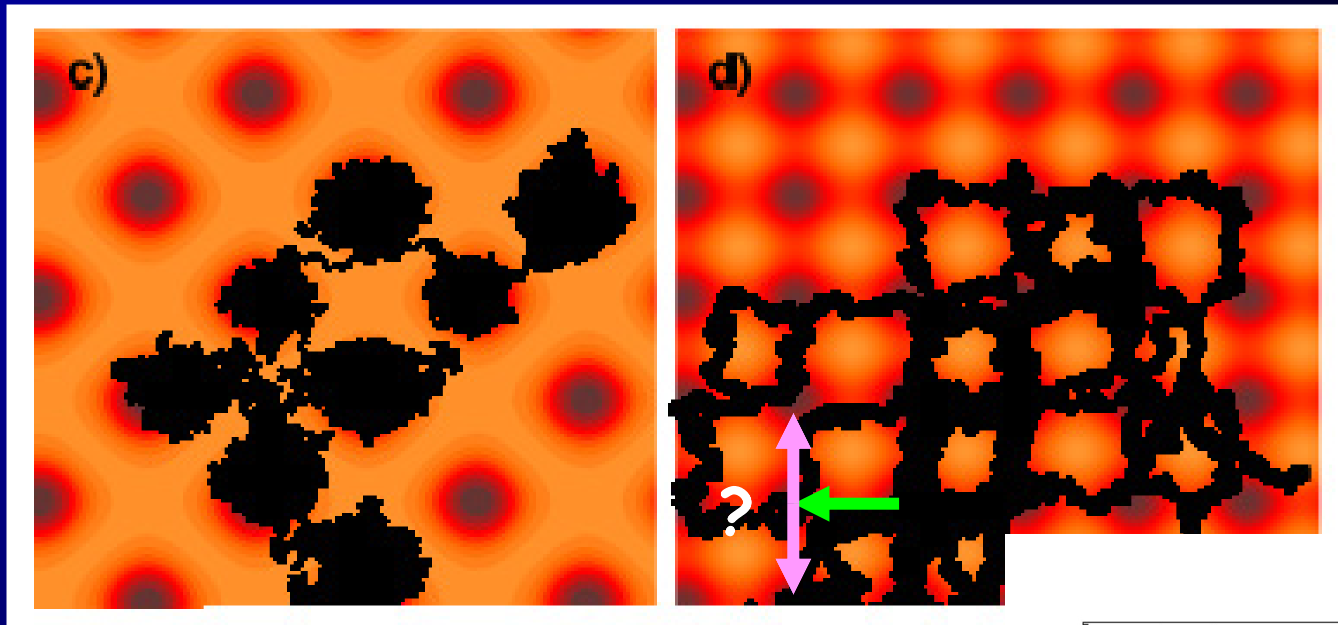
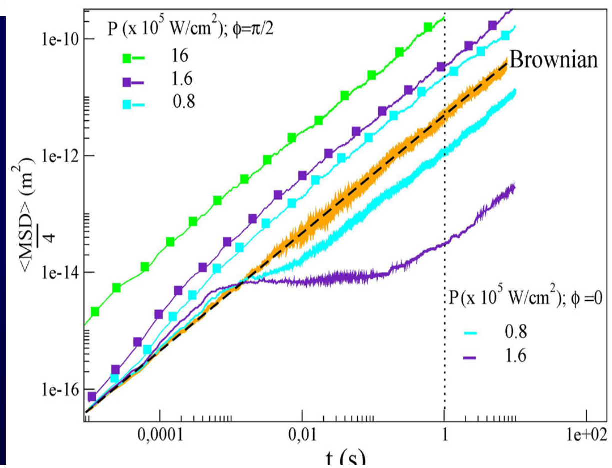


$$\tau \approx 10^{-5} - 10^{-4}\ \mathrm{s}$$

$$10^{6} - 10^{5}\ \ \mathrm{W/cm^{2}}.$$



$$D - D_0 \approx \left(\frac{\lambda}{2}\right)^2 \frac{1}{4\tau} \approx 2\frac{(n/c)\alpha'' P}{\gamma} = D_0 2\frac{(n/c)\alpha''}{k_B T} P$$

Microseres

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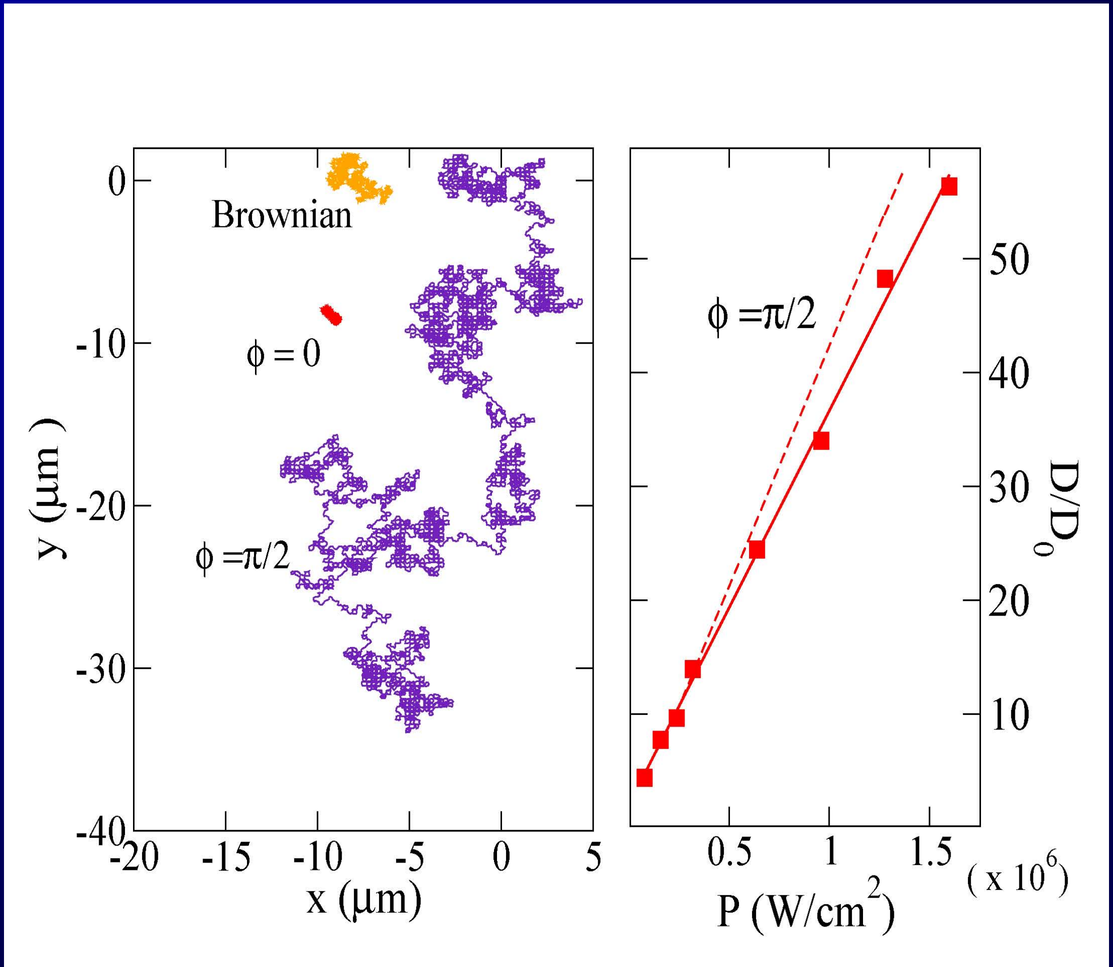
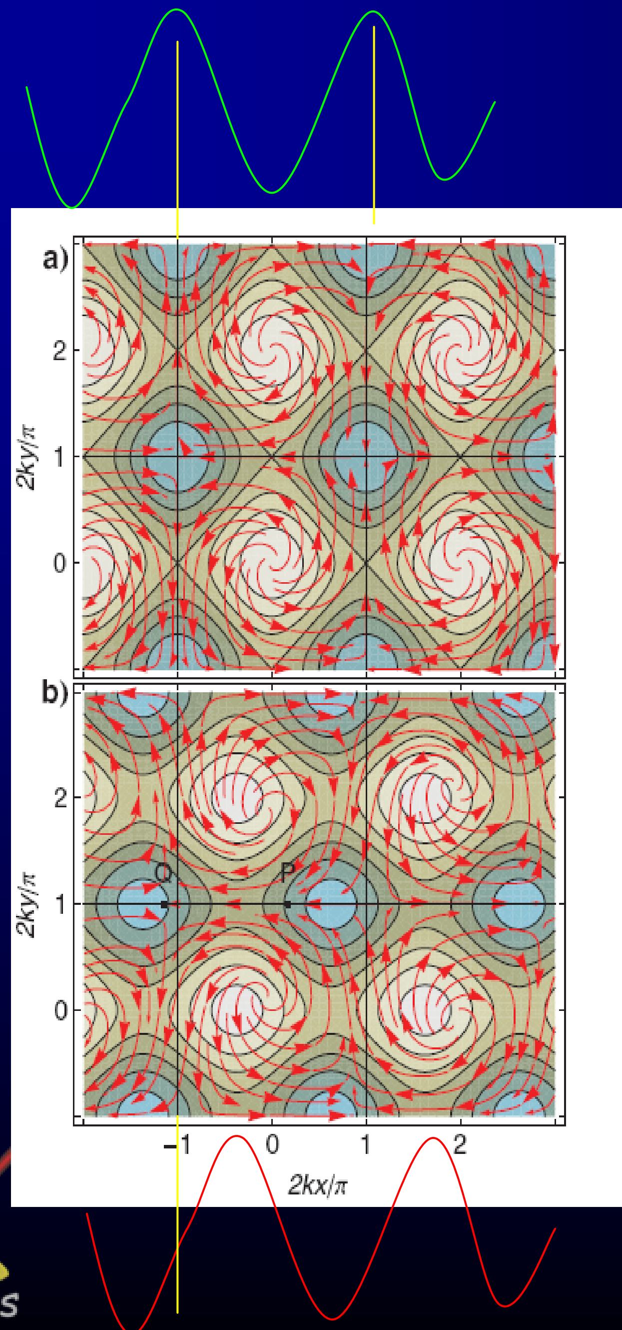
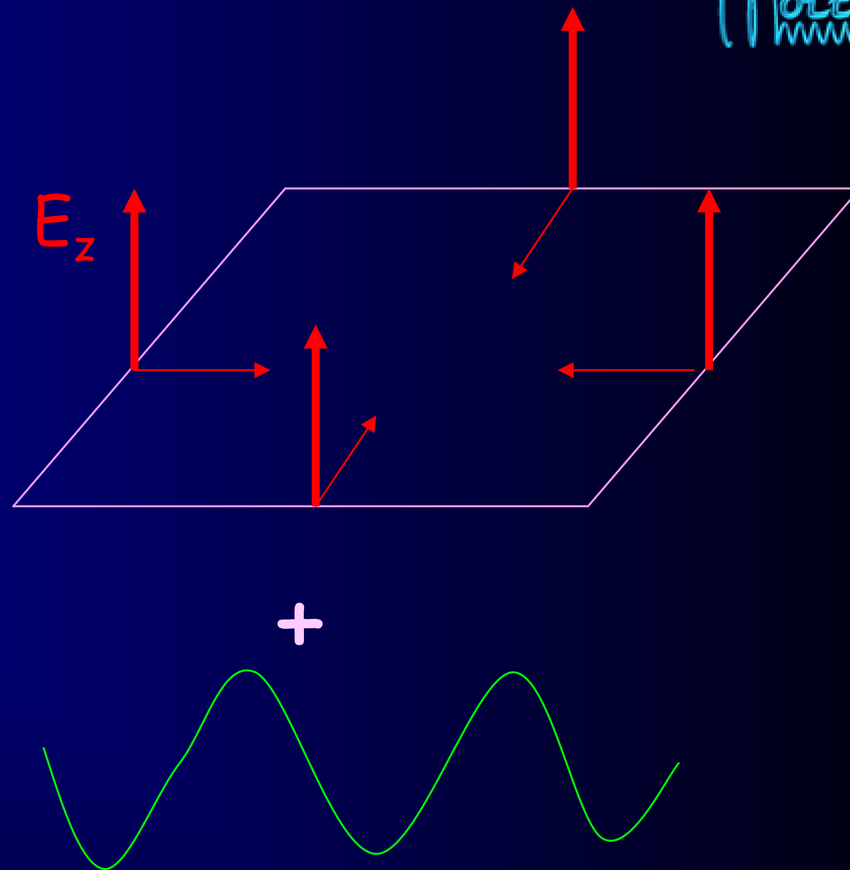


The diffusion constant of a 50 nm gold particle is enhanced by 2 orders of magnitude with respect to thermal diffusion at room temperature

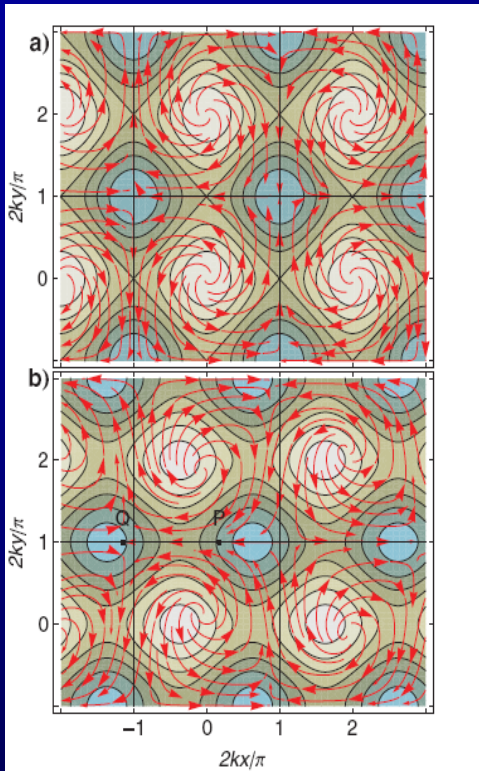
(S. Albaladejo et al. NanoLett. 2009)

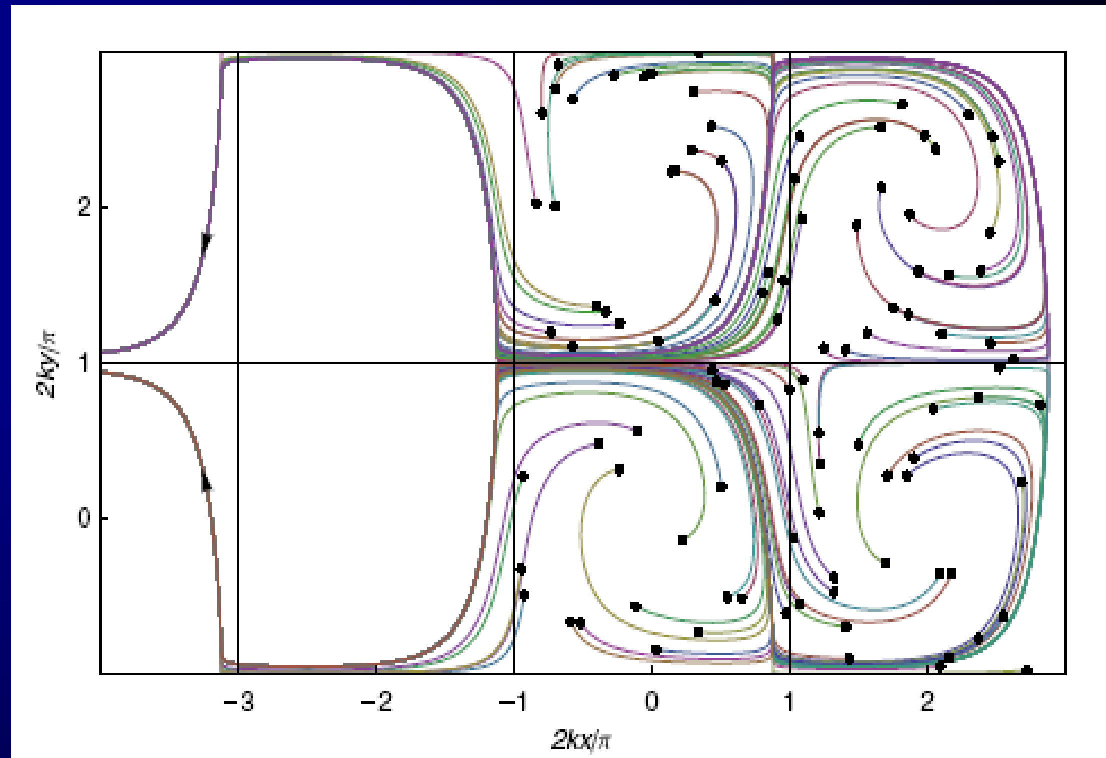




Deterministic Ratchet from Stationary Light Fields

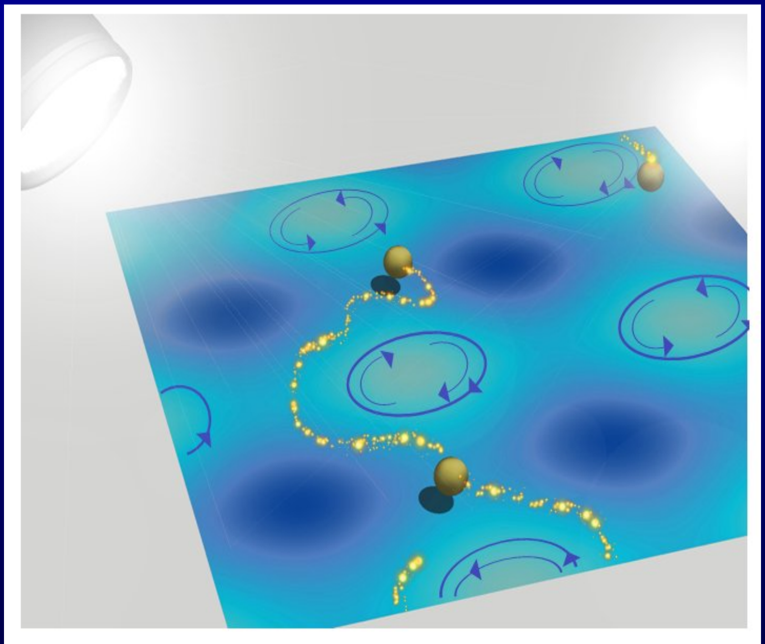
(I. Zapata et al. PRL 2009)

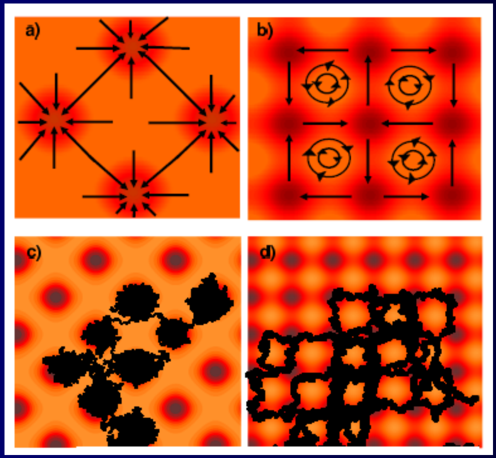


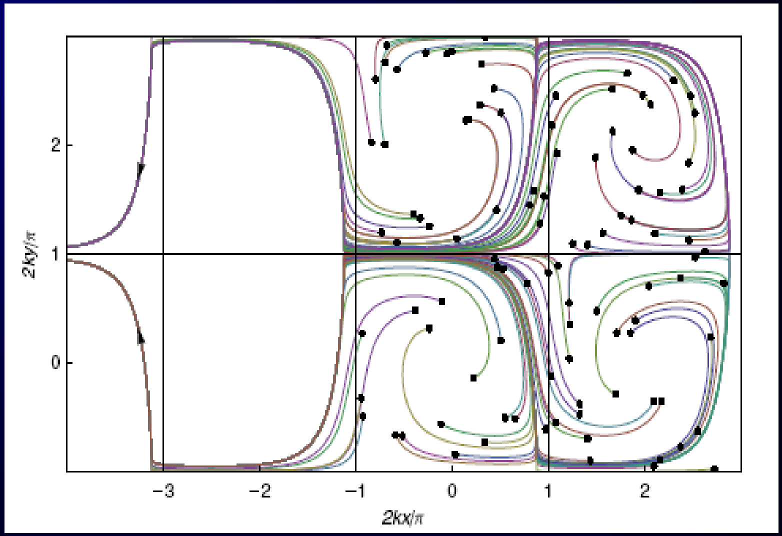


From stationary forces of null average we have designed a ratchet system which requires neither noise nor driving.

(I. Zapata etr al. PRL 2009)




a)
b)
c)
d)


2ky/π
2kx/π
−3
−2
−1
0
1
2