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Optical Tweezers on Nanostructures *Onofrio M. MARAGÒ*

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Laser On Stable trap <u>5 μm</u> onofrio.marago@cnr.it







Optical Tweezers People - Messina 2018



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Outline



- General introduction
- Theory background
 - ➤ T-matrix methods
- Experimental practice
- Linear nanostructures
 - Scaling, Binding & rotational dynamics
 - ➢ Air and vacuum
- Layered materials
- SERS Tweezers















J Kepler (1610) comet tails are the result of light pressure





> J C Maxwell (1864)

light pressure is explained in electromagnetic theory

- P Lebedev (1901) and Nichols & Hull (1901) measures light pressure for the first time
- Ashkin, T Haensch & A Schawlow, V Lethokov (1970s) first proposals to manipulate atoms and microparticles, laser cooling
- <u>A Ashkin & S Chu (1986)</u>

at Bell Laboratories moves and traps latex spheres suspended in water using a focused laser beam. Optical Tweezers are born!







Street Optical Trapping Theory

Optical trapping of particles is a consequence of the radiation force that stems from the **conservation of electromagnetic momentum in light scattering**.

Ray Optics, d/λ >> 1 Trapping forces from reflection and refraction of rays Forces proportional to gradient of intensity (Ashkin, Biophys. J. 61, 569, 1992)

Dipole Approximation, d/ $\lambda \ll 1$

Three parts: gradient force, scattering force (Ashkin, et al. Optics Lett. 11, 288, 1986)

Complex Region, $d/\lambda \approx 1$

- Full electromagnetic Theory (Maxwell ST)
- > Make use of **T-Matrix methods** for force & torque
- > Angular spectrum representation

Extension to complex non-spherical particles

$$\mathbf{F}_{\mathrm{Rad}} = r'^2 \int_{\Omega'} \mathbf{\hat{r}}' \cdot \langle \mathsf{T}_{\mathrm{M}} \rangle \,\mathrm{d}\Omega'$$

Nieminen&Rubinsztein-Dunlop,Gousbet,Neves,Hanna,Zemanek,...



 $\underline{F}_{\text{grad}} = -\underline{\nabla}U_{\text{dip}}$

 $\propto \underline{\nabla} I(\underline{r}) = -\kappa_i x_i$

Borghese, Denti, Saija, Springer (2007) Borghese et al., Optics Express (2007) Borghese et al., Phys Rev Lett (2008) Saija et al., Opt. Express (2009) ...









Diffusion Matrix Modeling for complex particles using Hydro++

G. Volpe & G. Volpe American Journal of Physics **81**, 224 (2013). A. Callegari, M. Mijalkov, et al. JOSA B **32**, B11-B19 (2015)

A. Stilgoe et al., Phys. Rev. Lett. 107, 248101 (2011).

By Magazzù, Callegari

Optical Tweezers – Dipole

Interaction of electric field of laser with induced dipole in dielectric $U = -p \cdot \underline{E} = -\alpha |E|^2$



Optical response of colloidal metal nanoparticles





d=30 nm Extinction cross-section (μm^2) Gold 0.015-Silver Aluminum 0.010-0.005 000. 200 300 400 500 600 100 700 Wavelength λ (nm)

Amendola, V., et al. (2017). Surface plasmon resonance in gold nanoparticles: a review. *J. Phys.: Cond. Matt.*, **29**, 203002. For **metal nanoparticles**, the presence of **plasmon resonances** leads to their optical trapping with a wavelength in the **red side** of the spectrum.



Saija R et al. Optics Express (2009) Jones et al., ACS Nano (2009) – Au Nanorods Messina et al., Optics Express (2015) – Ag Platelets

Optical trapping of plasmonic particles (dipole picture)



Lorentz-Drude model for dielectric constant using tabulated fit parameters that agrees well with experimental data and exploit plasmon resonance to enhance optical forces

$$\varepsilon(\omega) = \varepsilon_{\infty} + \sum_{k=1}^{K} \frac{f_k \omega_p^2}{\omega_k^2 - \omega^2 + i\omega\Gamma_k}$$

$$\alpha(\omega) = 4\pi\varepsilon_0 a^3 \frac{\varepsilon_1(\omega) - \varepsilon_2}{\varepsilon_1(\omega) + 2\varepsilon_2}$$

$$\sigma_{abs} = \frac{k}{\varepsilon_0} \operatorname{Im}\{\alpha(\omega)\}; \quad \sigma_{scat} = \frac{k^4}{6\pi\varepsilon_0^2} |\alpha(\omega)|^2$$

$$F_{scat}(r) = \frac{n \sigma_{ext}}{c} I(r)$$
$$F_{grad} = \frac{n}{2} \frac{\text{Re}\{\alpha(\omega)\}}{c \varepsilon_0} \nabla I(r)$$

nn



Trap Stiffness proportional to $Re\{\alpha(\omega)\}$

Trapped resonant gain metal/dielectric nanoshell



Brownian Dynamics Simulation

Overdamped $\frac{d}{dt}r(t) = -\frac{1}{\gamma}\frac{d}{dr}U(r) + \xi(t) \,.$ Langevin equation G. Volpe and G. Volpe, Integrating Am. J. Phys. (2013) $\begin{cases} x_i = x_{i-1} - \frac{\kappa_x}{\gamma} x_{i-1} \Delta t + \sqrt{2D\Delta t} w_{x,i} \\ y_i = y_{i-1} - \frac{\kappa_y}{\gamma} y_{i-1} \Delta t + \sqrt{2D\Delta t} w_{y,i} \end{cases}$ P = 20 mW $z_i = z_{i-1} - \frac{\kappa_z}{\gamma} z_{i-1} \Delta t + \sqrt{2D\Delta t} w_{z,i}$ t = 1 ms $\Delta t = 2 \text{ ns}$ G = -0.022G = -0.132G = -0.22 $\times 10^5$ 0.5 0.5 0.5 12 - displacement (µm) - displacement (µm) displacement (µm) 10 8 0 6 4 > > 2 -0.5 -0.5 -0.5 0 0 0.5 0.5 -0.5 0 0.5 -0.5 -0.5z - displacement (μ m) z - displacement (μ m) z - displacement (μ m)

P. Polimeno, A. Veltri, submitted

Optomechanical Position Locking and Channelling





P. Polimeno, A. Veltri, submitted



Nieminen, JQSRT (2003-2016); Singer et al., PR E (2006); Borghese, Opt. Express (2007); Borghese, Phys Rev Lett (2008); Saija, et al., Opt. Express (2009); Bareil & Sheng, Opt. Express (2010); Simpson & Hanna, PR E (2010); Simpson, JQSRT (2016);...











See also Saija et al. (2005), Borghese et al. (2006), Borghese et al. (2007)













Experimental practice



G. Pesce, et al, Step-by-step guide to the realization of advanced optical tweezers, *JOSA B* (2015) Special Issue Opex & JOSA B on Optical Cooling & Trapping

Holographic Optical Tweezers





 $\mathbf{E}_{0,0}^{\mathrm{L}\,\mathrm{G}}$

 $\mathbf{E}_{0,1}^{LG}$

 $\mathbf{E}_{0,2}^{LG}$

Laguerre-Gauss Beams can transfer Orbital Angular Momentum



 2π

Phase Mask



 π





Jones, Maragò, Volpe, "Optical Tweezers", CUP (2015) G. Pesce, et al, JOSA B (2015)



Advanced Optical Tweezers (Speckle fields)



Optical sorting with speckle fields

High intensity traps large particles, while small particles move in the microfluidic flow





Jones, Maragò, Volpe, "Optical Tweezers", CUP (2015) G. Pesce, et al, JOSA B (2015)

PCF OT of Linear Nanostructures





Nanotubes:

Tan et al., Nano Lett. (2004) Plewa et al., Optics Express (2004) Zhang et al., APL (2006) <u>O.M. Maragò</u> et al., Physica E (2008) <u>O.M. Maragò</u> et al., Nano Lett. (2008) <u>P.H. Jones</u>, et al. ACS Nano (2009) Pauzauskie et al, APL (2009) <u>Donato</u> et al., Opt. Lett. (2012)

> Nanofibers: <u>Neves</u> et al., Optics Express (2010)





Nanowires:

Agarwal et al., Optics Express (2005) Pauzauskie et al., Nat. Mat. (2006) Nakayama et al., Nature (2007) <u>Borghese</u> et al., Phys. Rev. Lett. (2008) Carberry et al., Nanotech. (2010) Simpson&Hanna, JOSA A (2010) Simpson&Hanna, PR E (2010) Reece et al., Nano Lett. (2011) Dutta et al., Nano Lett. (2011) <u>Irrera</u> et al., Nano Lett. (2011)








Brownian Motion of Nanotubes/Nanowires Shape determines the effective potential



O. M. Maragò, Nano Letters 8, 3211–3216 (2008); A. Irrera, Nano Letters 11, 4879–4884 (2011).

OT of Silicon Nanowires

A. Irrera et al., *Nano Letters* **2011**, *11*, 4879 A. Irrera, A. Magazzù, et al., *Nano Letters* **2016**.



We can now control the length and the diameter

Length controls optical forces and torques

Size-scaling with the size parameter xL=πnL/λ



Optical trapping of SiNW with controlled size



High resolution photonic force microscopy (F. Pedaci, CNRS, Montpellier)



Nanofabricated birifrengent particles with nanometric needles





Accurate force calibration of non-spherical probes



Topography of soft materials Membrane of <u>living</u> malaria-infected red blood cells presenting knobs





Pedaci et al. Nat. Phys (2010); Desgarceaux et al. arXiv:2002.01533 (2020)



Silicon nanowires in 2-beam traps Rich scenario regulated by shape & polarization θ_{y} Parallel linear polarization (PLP)

b)

c)

d)

[m]202

50

0

150

time [ms]

100

200

250

300

leads to **several equilibrium configuration** regulated by the **length** of the NWs

The NW scatters "mainly" from endtips

Polarization can be used to control orientation





By changing the size, w_0 , of the laser beams we can control the distance and binding interaction









Layered materials





D

1200

G

Intensity (arb. un.)

Raman Tweezers

- Lamp • 561nm,633nm,785nm... Excitation/trapping beam Edge/Notch filter Raman signal Condenser Illumination Sample Jobin-Yvon Triax 190 PI Stage spectrometer 100X Beam 1.3 NA • Avalanche photodiode, SPC Expander Laser HeNe Mirror (Perkin Elmer) Interference Filter **Raman & Photoluminescence** Edge inspection of trapped samples Filter E.g., Inspection of **Graphene** flakes Video Camera **Beam Splitter** Trapped Flake Spectrometer (exc. 633 nm) Avalanche 2D **Photodiode** Mirror D' D+D' Pinhole 2D'. 2000 2400 1600 2800 3200 Maragò, et al., "Brownian Motion of Graphene", ACS Nano 4, 7515 (2010) Raman shift (cm¹) Maragò, et al. Nature Nanotechnology 8, 807–819 (2013)





- hBN flakes trapped with 785 & 830 nm laser
- Trapped flakes have a range of peak positions. The blue-shifted positions (4cm⁻¹) of some peaks (~1370cm⁻¹) suggests that monolayer flakes are present in the sample. Presence of bi-layers (that should show a 2 cm⁻¹ red shift) is also evident.

Donato et al., Nanoscale (2018)

PCF Optical force calibration of hBN

flake

size



Optical force scaling on hBN

Two-dimensional scaling of optical trapping forces

Polarizability depends on the **area** of the layered particle

Flattening for large flakes (dipole approximatin breaks down)





Optical force positioning of MoS2 & WS2



Begin pushing

LG-Beam (I=30)

After few minutes





start







end

Use proteins (BSA) to glue the structures to a simple glass substrate



Raman Tweezers for Small Microplastics and Nanoplastics Identification in Seawater

www.acs.org





Raman Tweezers permits one to assess the **size and shape** of particles (beads, fragments, and fibers), with spatial resolution only limited by diffraction

Gillibert, R., et al. *Env. Science & Technol.* **53** (2019): 9003.







SERS Tweezers

SERS phenomenology

- 1. Molecules laying on metal nanoparticles and in their interstices <u>experience an</u> <u>enhanced excitation field</u> due to localized plasmon resonances
 - 2. Molecules will experience an <u>enhancement of the scattered fields</u>









Use the <u>SAME</u> light to push and excite SERS, Pushing wavelength 633nm < 687nm Nanorods SPR Proteins (BSA) are detected directly in their natural (PH) liquid environment with high sensitivity B. Fazio, et al., Scientific Reports 6 (2016): 26952





SERS Detection at low molar concentration



Creation of HOT SPOT region in liquid environment for high sensitive spectroscopy







Controlled patterning for in situ SERS detection





A. Foti et al., Materials 11, 440 (2018)



Optical forces were "born" in SPACE



Starshot project for a laser-driven lightsail



Nanomaterials and nanophotonic design



Atwater et al. Nat Mater. (2018)

Bring them back "home"

Trap and investigate **nanoparticulate** matter in space or planetary atmospheres



Thank You! Sunset from Vulcano, Onofrio M. Maragò

SPACE Tweezers Optical forces were "born" in SPACE



Starshot project for a laser-driven lightsail

Bring them back "home"

Trap and investigate nanoparticulate matter in space or planetary atmospheres





Localised surface plasmon





⇒ Collective oscillation of the electrons inside the nanoparticle ⇒ Interaction with light

Amendola, V., Pilot, R., Frasconi, M., Maragò, O. M., & Iatì, M. A. (2017). Surface plasmon resonance in gold nanoparticles: a review. *J. Phys.: Cond. Matt.*, *29*, 203002.





- Protein-Nanorods complexes in solution are composed by individual NRs surrounded by protein layer.
- Broadening and red-shift of the plasmon resonance, nanorods are coupled. This leads to enhanced fields, and then, SERS amplification of biomolecules spectra.

B. Fazio, et al., Scientific Reports (2016)
STPCF Optical Tweezers & Force Sensing



- Standard OT with QPD forward or back detection
- Multiwavelength: 830nm, 785nm, 633nm,

417nm, White Light Source

- Radial Polarizer (arcoptics)
- Piezostage (1nm resolution)
- LC waveplate
- Galvomirrors

Back focal plane interferometry combined with a QPD is sensitive to Brownian fluctuations

Brownian motion is a key ingredient in Force Sensing with optical tweezers.

NIR light ensures very low water absorption





EXAMPLE 1 Equation of motion of a damped harmonic oscillator subject to a randomly fluctuating force:

$$m_{dt}^{2} + \sqrt{\frac{dx}{dt}} + \sqrt{\frac{cx}{dt}} + \xi(t)$$
Stokes Trap Stokes Trap

$$\langle \xi(t) \rangle = 0$$
 $\langle \xi(t+\tau)\xi(t) \rangle = \frac{2\kappa_B T}{\gamma} \delta(\tau)$

• Equation of motion in the overdamped regime:

$$\gamma \partial_t x(t) = -\kappa x(t) + \xi(t)$$

Calculate the autocorrelation of position fluctuations:

$$C_{xx}(\tau) = \langle x(t)x(t+\tau) \rangle$$

The solution to which is straightforward:

$$C_{xx}(\tau) = C_{xx}(\tau = 0) \exp(-\omega\tau)$$

$$\omega = \frac{\kappa}{\gamma}$$











Brownian Motion is more complex

$$\partial_t X_i(t) = -\omega_i X_i(t) + \xi_i(t), \quad i = x, y, z$$

$$\partial_t \Theta_j(t) = -\Omega_j \Theta_j(t) + \xi_j(t), \quad j = x, y$$

From correlation functions we can extrapolate the <u>force</u> and <u>torque</u> constants on the SWNT bundle

$$C_{X_iX_i}(\tau) = \langle X_i(t)X_i(t+\tau) \rangle$$

$$C_{\Theta_j\Theta_j}(\tau) = \langle \Theta_j(t)\Theta_j(t+\tau) \rangle$$

$$\begin{split} \omega_x &= \Gamma_{\perp} k_x, \ \omega_y = \Gamma_{\perp} k_y, \ \omega_z = \Gamma_{\parallel} k_z \\ \Omega_x &= \Gamma_{\Theta} k_{\Theta_x}, \ \Omega_y = \Gamma_{\Theta} k_{\Theta_y}. \end{split}$$

Relaxation Frequencies for Translational and Angular Motion

Hydrodynamics of a rod-like nanostructure is embedded in the relaxation frequencies

Surface Plasmon Polaritons and Localized Surface Plasmons



Amendola, V., Pilot, R., Frasconi, M., Maragò, O. M., & Iatì, M. A. (2017). Surface plasmon resonance in gold nanoparticles: a review. *J. Phys.: Cond. Matt.*, *29*(20), 203002.



SERS enhancement due to **AuNPs in the inner walls** of Mesocapsule, that can be reached by MB molecules thanks to the **porosity of the shell**.

Optical Trapping in Dipole Approximation

- > Particle size parameter is small, $x = k_m a \ll 1$
- > Interaction of electric field of laser with induced dipole in dielectric $\mathbf{p}(\mathbf{r},t) = \alpha_{p} \mathbf{E}(\mathbf{r},t)$
- > The (harmonic) trapping potential is defined by the incident light intensity

$$\langle \mathbf{F} \rangle_{\mathrm{DA}} = \frac{1}{2} \frac{n_{\mathrm{m}}}{c \varepsilon_{\mathrm{m}}} \Re \left\{ \alpha_{\mathrm{p}} \right\} \nabla I(\mathbf{r}) + \frac{n_{\mathrm{m}}}{c} \phi_{\mathrm{r}}(\mathbf{r}) \\ \mathbf{f}_{\mathrm{c}} \phi_{\mathrm{r}}(\mathbf{r}) = -\underline{p} \cdot \underline{E} \\ \underline{F}_{\mathrm{grad}} \propto \underline{\nabla} I(\underline{r}) = -\kappa_{i} x_{i}$$
 (r)

J. R. Arias-González and M. Nieto-Vesperinas, JOSA A (2003)

Counter-propagating Gaussian beam

$$\kappa_{\rho}^{\text{c.p.}} = 8 \frac{\Re \{\alpha_{\rm p}\}}{cn_{\rm m}} \frac{I_0}{w_0^2}. \qquad z_0 = \frac{k_{\rm m} w_0^2}{2} \qquad w_0 = 0.5 \ \lambda_0 / NA$$
$$\kappa_z^{\text{c.p.}} = 4 \frac{\Re \{\alpha_{\rm p}\}}{cn_{\rm m}} \left(2 - 2k_{\rm m} z_0 + k_{\rm m}^2 z_0^2\right) \frac{I_0}{z_0^2}. \qquad I_0 = 2P / \pi w_0^2$$

O. Brzobohatý et. al. *Opt. Exp.* (2015)



Paolo Polimeno

Resonant gain metal/dielectric nanoshell



A. Veltri and A. Aradian, Phy. Rev. B (2012); A. Veltri et al., Sci. Rep. (2016)





F. Borghese, P. Denti, and R. Saija, Scattering from model non-spherical particles (Springer, 2007)



The Scattering Problem – Transition Matrix

Expansion of the scattered wave

$$\mathbf{E}_{s}(r,\hat{\mathbf{r}}) = E_{i} \sum_{l=0}^{\infty} \sum_{m=-l}^{l} A_{s,lm}^{(1)} \mathbf{H}_{lm}^{(1)}(r,\hat{\mathbf{r}}) + A_{s,lm}^{(2)} \mathbf{H}_{lm}^{(2)}(r,\hat{\mathbf{r}})$$

Expansion of the internal field

$$\mathbf{E}_{\mathbf{p}}(r,\hat{\mathbf{r}}) = E_{\mathbf{i}} \sum_{l=0}^{\infty} \sum_{m=-l}^{l} W_{\mathbf{p},lm}^{(1)} \mathbf{J}_{lm}^{(1)}(r,\hat{\mathbf{r}}) + W_{\mathbf{p},lm}^{(2)} \mathbf{J}_{lm}^{(2)}(r,\hat{\mathbf{r}})$$

Boundary conditions

$$\hat{\boldsymbol{n}} \times (\boldsymbol{E}_2 - \boldsymbol{E}_1) = 0$$

By imposing the Boundary conditions at the particle surface it is possible to find the relation between the **A** and **W** coefficients

$$A_{s,l'm'}^{(p')} = \sum_{p=1,2} \sum_{l=0}^{\infty} \sum_{m=-l}^{l} \underbrace{T_{lml'm'}^{(pp')}}_{i,lm} W_{i,lm}^{(p)}$$

 $\mathbf{E}_{s} = \mathbb{T}\mathbf{E}_{i}$ T- Matrix

Borghese et al., J. Math. Phys. (1980); Borghese et al. Aerosol Sci. & Tech. (1984)

ZnO nanowires in 2-beam traps in air and vacuum





Commercial samples L=1 micron, d=70 nm

NA=0.5, $w_0 = 2.8 \,\mu\text{m}$ beam waist





Damping decreases with pressure, hence rotational frequency increases Length controls both transferred torque and damping