Optical Tweezers on Nanostructures

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Optical Tweezers People - Messina 2018

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Acknowledgements

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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
Light Moves Matter

- 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

- A Ashkin, T Haensch & A Schawlow, V Lethokov (1970s)
  first proposals to manipulate atoms and microparticles, laser cooling

- A Ashkin & S Chu (1986)
  at Bell Laboratories moves and traps latex spheres suspended in water using a focused laser beam. **Optical Tweezers** are born!
THE NOBEL PRIZE IN PHYSICS 2018

Arthur Ashkin  Gérard Mourou  Donna Strickland

“for groundbreaking inventions in the field of laser physics”

THE ROYAL SWEDISH ACADEMY OF SCIENCES
Celebrations for Arthur Ashkin’s Nobel prize 2018
Optical Trapping at the Nanoscale

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

Ray Optics, \( d/\lambda \gg 1 \)
- 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

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

Borghese et al., Optics Express (2007)
Saija et al., Opt. Express (2009) …
Optical Tweezers - Ray Optics

Microsphere acts as a lens for the refracted and reflected rays

Linear momentum exchange from light to particle, push towards beam center \((n_p > n_m)\)

Trapping Force is proportional to the gradient of light intensity

Scattering force is proportional to light intensity and directed as the Poynting vector

\[
F = \sum_m F^{(m)} = \sum_m \left[ \frac{P_i^{(m)}}{c_i} \hat{u}_i^{(m)} - \frac{P_r^{(m)}}{c_i} \hat{u}_r^{(m)} - \sum_{n=1}^{+\infty} \frac{P_{t,n}^{(m)}}{c_i} \hat{u}_{t,n}^{(m)} \right]
\]
Dynamics of a spheroid in a double trap

Diffusion Matrix Modeling for complex particles using Hydro++

Optical Tweezers – Dipole

Interaction of electric field of laser with induced dipole in dielectric: \( U = -p \cdot E = -\alpha |E|^2 \)

Average force on particle: \( \langle F \rangle = \frac{1}{2} \text{Re} \left( \sum_i \alpha E_i \nabla E_i^* \right) \)

For a Gaussian beam breaks in two contributions:

- Scattering force: \( F_{\text{scatt}} = \frac{\hbar k}{2} \frac{I/I_{\text{sat}}}{1 + I/I_{\text{sat}} + 4 \delta^2 / \Gamma^2} \)
- Dipole force: \( F_{\text{dipole}} = -\frac{\hbar \delta}{2} \frac{\nabla (I/I_{\text{sat}})}{1 + I/I_{\text{sat}} + 4 \delta^2 / \Gamma^2} \)

The (harmonic) trapping potential is defined by the incident light intensity:

\( U_{\text{dip}} = -p \cdot E \)

\( F_{\text{grad}} = -\nabla U_{\text{dip}} \)

Optical response of colloidal metal nanoparticles

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

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

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

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

\[
F_{scat}(r) = \frac{n\sigma_{ext}}{c} I(r)
\]

\[
F_{grad} = \frac{n}{2} \frac{\text{Re}\{\alpha(\omega)\}}{c\varepsilon_0} \nabla I(r)
\]

Trap Stiffness proportional to \(\text{Re}\{\alpha(\omega)\}\)
Trapped resonant gain metal/dielectric nanoshell

Molecules

Nanoshell

Silver nanoshell with externally pumped optical gain material on its core excited by an external electric field.

Steady State Gain dielectric permittivity

\[ \varepsilon_1 = \varepsilon_b - \frac{G \Delta}{2(\omega - \omega_{21}) + i\Delta} \]

\[ \Delta = \frac{2}{\tau_2} \]

\[ \varepsilon_b \] dielectric host permittivity

\[ G = \Im[\varepsilon_1(\omega_{21})] = -\frac{n\mu^2\tau_2}{3\hbar\varepsilon_0} \tilde{N} \]

P. Polimeno, A. Veltri, submitted
Brownian Dynamics Simulation

Overdamped Langevin equation

\[
\frac{d}{dt} r(t) = -\frac{1}{\gamma} \frac{d}{dr} U(r) + \xi(t)
\]


\[
P = 20 \text{ mW} \\
t = 1 \text{ ms} \\
\Delta t = 2 \text{ ns}
\]

\[
G = -0.022 \\
G = -0.132 \\
G = -0.22
\]

P. Polimeno, A. Veltri, submitted
Optomechanical Position Locking and Channelling

\[ G = -0.22 \]
\[ P = 20 \text{ mW} \]
\[ t = 1 \text{ ms} \]
\[ \Delta t = 2 \text{ ns} \]

P. Polimeni, A. Veltri, submitted
Optical trapping of particles is a consequence of the conservation of electromagnetic momentum and the spatial redistribution of photons in light scattering.

**Mesoscopic Region, d/\lambda \approx 1**
- Full electromagnetic Theory
- Vector character of laser field
- Make use of Transition-Matrix approach
- Extension to non-spherical particles!

Conservation of linear momentum
\[
\left\langle \frac{dP_{Mech}}{dt} \right\rangle = \mathbf{F}_{Rad} = \oint_S \mathbf{n} \cdot \left\langle T_M \right\rangle dS
\]

Conservation of angular momentum
\[
\left\langle \frac{dL_{Mech}}{dt} \right\rangle = \Gamma_{Rad} = -\oint_S \mathbf{n} \cdot \left\langle T_M \right\rangle \times \mathbf{n} dS
\]

Averaged Maxwell stress tensor
\[
\left\langle T_M \right\rangle = \frac{1}{2} \varepsilon_m \mathfrak{R} \left\{ \mathbf{E}_t \otimes \mathbf{E}_t^* + \frac{c^2}{n_m^2} \mathbf{B}_t \otimes \mathbf{B}_t^* - \frac{1}{2} \left( \mathbf{E}_t^2 + \frac{c^2}{n_m^2} \mathbf{B}_t^2 \right) I \right\}
\]

Special case: Mie Scattering

Boundary conditions on a sphere
Only $l$-dependence
T-matrix is diagonal

Radiation pressure (plane wave)

$$
F_{\text{rad}} = \frac{n m}{c} I_i \left[ \sigma_{\text{ext}} - g_i \sigma_{\text{scat}} \right] \hat{k}_i
$$

Debye (1909), Mishchenko (2001)

Observables can be obtained from the Mie coefficients

$$
\sigma_{\text{ext}} = \frac{2\pi}{k_m^2} \sum_{l=1}^{\infty} (2l + 1) \text{Re} \left\{ a_l + b_l \right\}
$$

$$
\sigma_{\text{scat}} = \frac{2\pi}{k_m^2} \sum_{l=1}^{\infty} (2l + 1) \left( |a_l|^2 + |b_l|^2 \right)
$$

$$
g_i = \frac{4\pi}{\sigma_{\text{scat}} k_m^2} \text{Re} \left\{ \sum_{l=1}^{+\infty} \left[ \frac{l(l+2)}{l+1} (a_l a_{l+1}^* + b_l b_{l+1}^*) + \frac{2l+1}{l(l+1)} a_l b_l^* \right] \right\}
$$

T-matrix & Brownian dynamics
Modelling feedback cooling

Cooling in phase-space (feedback on x)

Feedback off-on

\[ F_{FB} = \beta \frac{r v_r}{\langle r \rangle_{\beta=0} \langle v_r \rangle_{\beta=0}} F_x \]


Extension to non-spherical particles
Mechanical effects on non-spherical particles

 Radiation Pressure
 Transverse Force \) \rightarrow \) Conservation of linear momentum
 Radiation Torque \) \rightarrow \) Conservation of angular momentum

 Radiation cross-sections yield the mechanical effects of light on non-spherical particles

 Wind sailing
 Solar sailing

 Swartzlander, Optical lift, Nature Photonics 2010
 Simpson et al, Opt Lett 37, 2012
Radiation force & torque from a general scattering process

Borghini equations

\[ F_{\text{rad}}(\hat{u}) = -\frac{\varepsilon_m E_i^2}{2k_m^2} \text{Re} \left\{ \sum_{plm} \sum_{p'l'm'} i^{-l'-l} I_{lml'm'}^{(pp')} (\hat{u}) \left[ A_{s,lm}^{(p)*} A_{s,l'm'}^{(p')} + W_{i,lm}^{(p)*} A_{s,l'm'}^{(p')} \right] \right\} \]

\[ T_{\text{rad},z} = -\frac{\varepsilon_m E_i^2}{2k_m^3} \sum_{plm} m \text{Re} \left\{ W_{i,lm}^{(p)} A_{s,lm}^{(p)*} \right\} - \frac{\varepsilon_m E_i^2}{2k_m^3} \sum_{plm} m |A_{s,lm}^{(p)}|^2 \]

- **Extension to hybrid structures:** See Ridolfo et al. ACS Nano (2011)
- **Extension to surfaces:** See Denti et al. JOSA A (1999).

See also Saija et al. (2005), Borghese et al. (2006), Borghese et al. (2007)
Size scaling for polystyrene spheres and nanowires

\[ Q_i = F_i \frac{c}{n_m P_i} \]

\[ \lambda = 830 \text{ nm} \]
\[ NA = 1.3 \]
\[ w_0 \approx 320 \text{ nm} \]

Sphere of radius \( a = 300 \text{ nm} \)

Nanowire of half-length \( L/2 = 300 \text{ nm} \)

\[ F_i = -\kappa_i x_i \]
Harmonic force trapping

Size scaling for a sphere

(a)

(b)

Size scaling for polystyrene spheres and nanowires

\[ \lambda = 830 \text{ nm} \]
\[ \text{NA} = 1.3 \]
\[ w_0 \approx 320 \text{ nm} \]
Scaling of Force Constants

T-matrix calculations – Role of shape

Maximum in the axial (z) force constant

Optimum overlap between wire and high intensity region (Rayleigh Range)

See also Simpson & Hanna, Nanotechnology (2012)
Experimental practice
Building an Optical Tweezers

Holographic Optical Tweezers

Laguerre-Gauss Beams can transfer Orbital Angular Momentum

Phase Mask

Trapped particles


Optical sorting with speckle fields

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

Nanowires:
Agarwal et al., Optics Express (2005)
Pauzauskie et al., Nat. Mat. (2006)
Nakayama et al., Nature (2007)
Carberry et al., Nanotech. (2010)
Simpson&Hanna, JOSA A (2010)
Reece et al., Nano Lett. (2011)
Dutta et al., Nano Lett. (2011)
Irrera et al., Nano Lett. (2011)
...

Nanotubes:
Plewa et al., Optics Express (2004)
Zhang et al., APL (2006)
Pauzauskie et al, APL (2009)

Nanofibers:
Neves et al., Optics Express (2010)
OT of Nanofibers

Hydrodynamics of a rod-like nanostructure is anisotropic.

\[
\Gamma_\perp = \frac{\ln p + \delta_\perp}{4\pi\eta L}, \quad \Gamma_\parallel = \frac{\ln p + \delta_\parallel}{2\pi\eta L}, \\
\Gamma_\Theta = \frac{3(\ln p + \delta_\Theta)}{\pi\eta L^3}
\]

The signals from the QPD are a composition of center of mass \( X_i \) and angular motion \( \Theta_i \).

\[
S_x \sim \beta_x (X + a \Theta_x); \quad S_y \sim \beta_y (Y + b \Theta_y); \quad S_z \sim \beta_z Z
\]

Small angle approximation
Correlation functions of the tracking signals give information on torque and force constants.

\[ C_{ii}(\tau) = \langle S_i(t)S_i(t+\tau) \rangle \]
\[ C_{xy}(\tau) = \langle S_x(t)S_y(t+\tau) \rangle \]

- **Double Exp for “fast” dynamics**
- **Simple Exp for Z and Cross**

Non-conservative forces:
Brownian Motion of Nanotubes/Nanowires

Shape determines the effective potential

Transverse fluctuations in the 10 nm range

OT of Silicon Nanowires
A. Irrera et al., *Nano Letters* 2011, 11, 4879

We can now control the length and the diameter.

Length controls optical forces and torques.

Size-scaling with the size parameter $x_L = \frac{\pi n L}{\lambda}$

Optical trapping of SiNW with controlled size
Scaling of Force Constants

Experiment
Irrera et al., Nano Letters (2011)

T-matrix
By P. Polimeno

For long nanowires transverse force saturates, axial force drops.
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 living malaria-infected red blood cells presenting knobs

Silicon nanowires in 2-beam traps

Silicon nanowires in 2-beam traps
Rich scenario regulated by shape & polarization

Parallel linear polarization (PLP) leads to several equilibrium configuration regulated by the length of the NWs.

The NW scatters “mainly” from end-tips.

Polarization can be used to control orientation.

From PLP to XLP (fringes disappear) longitudinal torque wins over polarization torque.
By changing the size, $w_0$, of the laser beams we can control the distance and binding interaction.
Rich spin rotational dynamics regulated by shape

Imaging the XZ plane

Simulations by S. H. Simpson
Rich orbital rotational dynamics regulated by shape

Imaging the XZ plane

Sim. by S. H. Simpson

XY

XY

XY
Layered materials
Liquid Phase Exfoliation

Graphene

A

SDC

B

Graphite

C

Mild sonication

2d Materials Inks
Flexible electronics and textile

Raman Tweezers

- 561nm, 633nm, 785nm...
- Edge/Notch filter
- Jobin-Yvon Triax 190 spectrometer
- Avalanche photodiode, SPC (Perkin Elmer)

Raman & Photoluminescence inspection of trapped samples

E.g., Inspection of Graphene flakes

Mechanical effects of light on layered materials

Donato et al., Nanoscale (2018)
• hBN flakes trapped with 785 & 830 nm laser
• Trapped flakes have a range of peak positions. The blue-shifted positions (4 cm\(^{-1}\)) of some peaks (~1370 cm\(^{-1}\)) 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.
Optical force calibration of hBN

Microscope stage calibration combined with ACFs

Microscope stage oscillation yield a sinusoidal modulation of the particle’s displacement

\[ V_x(t) = x_V(t) + a_V \sin(\omega_{\text{stage}} t) \]

All parameters from ACF fitting

Hydrodynamics for a thin disk

\[
\begin{align*}
\gamma_{||} &= 8 \eta D \\
\gamma_{\perp} &= \frac{16}{3} \eta D \\
\gamma_{r} &= \frac{4}{3} \eta D^3
\end{align*}
\]

ACFs full calibration enable flake size estimates from a simple hydrodynamic model

Donato et al., Nanoscale (2018)
Two-dimensional scaling of optical trapping forces

Polarizability depends on the area of the layered particle

Flattening for large flakes (dipole approximation breaks down)

Donato et al., Nanoscale (2018)
Optical force positioning of MoS\textsubscript{2} & WS\textsubscript{2}

Begin pushing  
LG-Beam ($l=30$)  
After few minutes

Use proteins (BSA) to glue the structures to a simple glass substrate
Time-evolution of optical force aggregation of MoS2

Donato et al., Nanoscale (2018)
Raman Tweezers permits one to assess the size and shape of particles (beads, fragments, and fibers), with spatial resolution only limited by diffraction.

SERS phenomenology

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

\[ E_{loc} = \Gamma_{exc}(\lambda_L)E_0 \]

\[ E_{SERS} = \Gamma_{rad}(\lambda_R)E_{Raman} \]

OT of Au NanoAggregates

LASiS

I) 1064nm (9ns) Laser pulse
II) pyridine
III) bsa

Extinction (a.u.)

Messina et al., ACS Nano 5, 905 (2011)
Use the **SAME** light to trap and excite SERS
Trapping wavelength $785\text{nm} > 695\text{nm}$ MNP Aggregate SPR

The huge electromagnetic field enhancement enables dramatic increase of the vibrational signal of molecules located in the *hot spots.*
A biosensor based on optical forces

Optical forces aggregates plasmonic particles and create “hot spots” on demand

Low NA = 0.5-0.9

Use the **SAME** 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

Time evolution of SERS spectra

a. Phe ring breathing
b. Aromatic aminoacids and Amide I
c. CH stretching region

Time evolution of SERS signals can be used to study the dynamics of aggregation. This depends on wavelength and power.
SERS Detection at low molar concentration

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

- Detection of BSA at concentrations as low as $10^{-7} \text{ M}$
- Enhancement of BSA Raman scattering by 5 orders of magnitude

Results also with Lysozyme, Phenylalanine, and Mnsod

Wavelength dependence

P = 4 mW, after 500 s


a) \( \lambda = 650 \text{ nm} \)

b) \( \lambda = 700 \text{ nm} \)

Average velocity of aggregation is estimated in the linear regime (before saturation) as the size of the aggregate divided by the aggregation time.
Power dependence of aggregation velocity

Controlled patterning for in situ SERS detection

Optical forces were "born" in SPACE

Bring them back "home"
Trap and investigate nanoparticulate matter in space or planetary atmospheres

Starshot project for a laser-driven lightsail

Nanomaterials and nanophotonic design

Thank You!

Sunset from Vulcano, Onofrio M. Maragò
Optical forces were “born” in SPACE

SPACE Tweezers

Bring them back “home”

Trap and investigate nanoparticulate matter in space or planetary atmospheres

Starshot project for a laser-driven lightsail

Nanomaterials and nanophotonic design

Localised surface plasmon

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

Plasmonic Tweezers
Juan et al., Nat. Photon. 5, 349 (2011); Maragò et al., Nat. Nano. 8, 807 (2013)

Surface Plasmon Polaritons
Plasmonic landscape (microscale)

Localized Plasmon Polaritons
Nanoantennas (hot spot trapping, nanoscale)


BSA+Nanorods Extinction

In solution UV spectra do not change

NRs are aggregated by optical forces and glued by plasmonic heating at the surface of the sample chamber.

- **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.

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
How do we measure forces?

We measure the interference pattern between scattered and unscattered light in the back focal plane on a QPD. Time evolution is proportional to the particle positional fluctuations.

Brownian motion is the key ingredient to calibrate Optical Tweezers.

\[ S_\chi = (Q1 + Q3) - (Q2 + Q4) \]
Brownian Motion for a bead

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

  \[ m \frac{d^2 x}{dt^2} + \gamma \frac{dx}{dt} + \kappa x = \xi(t) \]

- The term \( \xi(t) \) describes random (uncorrelated) fluctuations in force with zero mean, i.e.

  \[ \langle \xi(t) \rangle = 0 \quad \langle \xi(t + \tau) \xi(t) \rangle = \frac{2 k_B T}{\gamma} \delta(\tau) \]

- 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:

  \[ C_{xx}(\tau) = C_{xx}(\tau = 0) \exp(-\omega \tau) \quad \omega = \frac{\kappa}{\gamma} \]
Calibration for a bead

From QPD tracking signals we get Autocorrelation Functions and eventually the Force Constants

\[
C_{xx}(\tau) = C_{xx}(0) \exp \left( -\frac{k_x}{\gamma} \tau \right)
\]

\[
C^V_{xx}(\tau) = \langle V_x(t)V_x(t+\tau) \rangle = \beta_x^2 C_{xx}(\tau)
\]

\[
C^V_{xx}(0) = \beta_x^2 C_{xx}(0) = \beta_x^2 \frac{k_B T}{k_x}
\]

Calibration factor

\[
\beta_x = \sqrt{\frac{C^V_{xx}(0) k_x}{k_B T}}
\]
Different Analysis for the same data

But different methods yield different information on the particle dynamics
From each signal we can reconstruct the effective trapping potential in 3D.
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 force and torque constants on the SWNT bundle.

\[ C_{X_iX_i}(\tau) = \langle X_i(t)X_i(t+\tau) \rangle \]
\[ C_{\Theta_j\Theta_j}(\tau) = \langle \Theta_j(t)\Theta_j(t+\tau) \rangle \]

\[ \omega_x = \Gamma_\perp k_x, \quad \omega_y = \Gamma_\perp k_y, \quad \omega_z = \Gamma_\parallel k_z \]
\[ \Omega_x = \Gamma_\Theta k_\Theta x, \quad \Omega_y = \Gamma_\Theta k_\Theta y. \]

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

Relaxation Frequencies for Translational and Angular Motion
Plasmonic Mesocapsules

**Mesocapsule:** external porous *Silica* shell 30\(\text{nm}\), internal *AuNPs* with diameters 11\(\text{nm}\). Capsule diameter 1.4 \(\text{microns}\).

![Mesocapsule image](image)

Cargo mesocapsules can be manipulated and activated to release specific molecules in-situ.


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*. 

![SERS Tweezers](image)
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 $p(r, t) = \alpha_p E(r, t)$
- The (harmonic) trapping potential is defined by the incident light intensity

$$\langle F \rangle_{DA} = \frac{1}{2} \frac{n_m}{\varepsilon_m} \Re \{ \alpha_p \} \nabla I(r) + \frac{n_m}{c} \nabla \Phi(r)$$

Gradient force

Scattering force


Counter-propagating Gaussian beam

$$\kappa_{\rho} = 8 \frac{\Re \{ \alpha_p \} I_0}{c n_m} \frac{l_0^2}{w_0^2}.$$  
$$z_0 = \frac{k_m w_0^2}{2}.$$  
$$w_0 = 0.5 \lambda_0 / NA.$$  
$$i_0 = 2P / \pi w_0^2.$$  


Paolo Polimenio
Resonant gain metal/dielectric nanoshell

Under a pumping threshold, nanoshell shows a stable dipolar field.

\[
\bar{p} = \frac{\alpha_{\text{NUM}(\varepsilon_1, \varepsilon_2, \varepsilon_3, \rho)}}{\alpha_{\text{DEN}(\varepsilon_1, \varepsilon_2, \varepsilon_3, \rho)}} \bar{E}
\]

\[\rho = \frac{a_1}{a_2}\]

\[a_1\text{ int. radius}\]
\[a_2\text{ ext. radius}\]

\[\varepsilon_3\text{ water (solvent) dielectric permittivity}\]

Steady State Gain dielectric permittivity

\[\varepsilon_1 = \varepsilon_b - \frac{G\Delta}{2(\omega - \omega_{21}) + i\Delta}\]

\[\Delta = \frac{2}{\tau_2}\]

\[\varepsilon_b\text{ dielectric host permittivity}\]

\[G = \Im[\varepsilon_1(\omega_{21})] = -\frac{n\mu_2^2\tau_2}{3}\tilde{N}\]

Single metallic nanoparticle permittivity (Drude model)

\[\varepsilon_2 = \varepsilon_\infty - \frac{\omega_p^2}{\omega(\omega + 2i\gamma)}\]

In our calculation,
\[\rho = 0.77\text{ and }a_2 = 20\text{ nm}\]

The Scattering Problem – Multipole Expansion

Helmholtz equations

\[
\left( \nabla^2 + n^2 k_v^2 \right) E = 0 , \quad \left( \nabla^2 + n^2 k_v^2 \right) B = 0
\]

Boundary conditions

\[
E_2 = E_i + E_s \quad E_1 = E_p
\]

\[
\hat{n} \times (E_2 - E_1) = 0 , \quad \hat{n} \times (B_2 - B_1) = 0
\]

Expansion of the incident field

\[
E_i(r, \hat{r}) = E_i \sum_{l=0}^{\infty} \sum_{m=-l}^{l} W^{(1)}_{i,lm} J^{(1)}_{lm}(r, \hat{r}) + W^{(2)}_{i,lm} J^{(2)}_{lm}(r, \hat{r})
\]

Magnetic multipole

\[
J^{(1)}_{lm} = j_l(kr) X_{lm}(\hat{r})
\]

Electric multipole

\[
J^{(2)}_{lm} = \frac{1}{k} \nabla \times J^{(1)}_{lm}
\]

Vector spherical harmonics

\[
X_{lm} = \left[ l(l + 1) \right]^{-1/2} L Y_{lm}
\]

The Scattering Problem – Transition Matrix

Expansion of the scattered wave

\[ E_s(r, \hat{r}) = E_i \sum_{l=0}^{\infty} \sum_{m=-l}^{l} A_{s,lm}^{(1)} H_{lm}^{(1)}(r, \hat{r}) + A_{s,lm}^{(2)} H_{lm}^{(2)}(r, \hat{r}) \]

Expansion of the internal field

\[ E_p(r, \hat{r}) = E_i \sum_{l=0}^{\infty} \sum_{m=-l}^{l} W_{p,lm}^{(1)} J_{lm}^{(1)}(r, \hat{r}) + W_{p,lm}^{(2)} J_{lm}^{(2)}(r, \hat{r}) \]

Boundary conditions

\[ \hat{n} \times (E_2 - E_1) = 0 \]

\[ E_2 = E_i + E_s \]

\[ E_1 = E_p \]

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

\[ E_s = TE_i \]

T- Matrix

ZnO nanowires in 2-beam traps in air and vacuum

Commercial samples
L=1 micron, d=70 nm

NA=0.5, $w_0 = 2.8 \mu m$ beam waist
QPD detection of rotational dynamics vs Pressure

Damping decreases with pressure, hence rotational frequency increases.

Length controls both transferred torque and damping.

Translational motion (background)

Rotational motion

\[
\text{PSD}(\Omega) = \frac{k_B T}{\pi m} \left[ \frac{\Gamma_{\text{transl}}}{(\Omega_{\text{transl}}^2 - \Omega^2)^2 + \Gamma_{\text{transl}}^2 \Omega^2} + \sum_i \frac{\Gamma_{\text{rot},i}}{(\Omega_{\text{rot},i}^2 - \Omega^2)^2 + \Gamma_{\text{rot},i}^2 \Omega^2} \right]
\]