Nonlinear transmission of light through synthetic colloidal suspensions

Zhigang Chen

San Francisco State Univ., California, USA
& Nankai Univ. China
What do we do with light?

- **Spatial solitons & dynamics** (PRL, OL, 2004-2009)
- **Condensed-matter photonics** (Nat Mat 2014, Nat Comm 2015)
- **Optical beam engineering** (PRL 2012, 2015, OL 2010-15)
- **Trapping and manipulation** (Opt Lett, OE, 2010-14)
- **Soft matter NLO** (PRL 2013, Nano Lett 2014)
Outline

• Motivation & background
  ➢ Synthetic colloidal suspensions
    (optical forces, polarizibility, tunable NL)
  ➢ Optofluidic manipulation, transport through scattering media…

• Dielectric suspensions
  ➢ Negative and mixed polarizibilities
  ➢ Self-induced transparency effects

• Metallic suspensions
  ➢ Tunable polarizabilities
  ➢ Plasmonic resonant solitons

• Biological suspensions
  ➢ Penetrating or killing?
  ➢ Nonlinear effects?
Optical nonlinearity in soft-matter systems - Motivation

An interdisciplinary field

Colloidal Physics
Soft-matter

Statistical Mechanics

Nonlinear Optics

Fluid mechanics

Chemistry/electrochemistry

Life sciences

Brownian motion
Optical forces
Nonlinear scattering
Soliton effects
Historical Overview


Nonlinear effects:

- **Four-wave mixing in artificial Kerr Media**

- **Self focusing in artificial Kerr media**

- **Soliton-like beams in aqueous suspensions**
  V.E. Yashin et al, Optics and spectroscopy (2005)

- **Optical Spatial Solitons in Soft Matter**

- **Soliton dynamics and self-induced transparency in nonlinear nanosuspensions**

- **Spatial solitons and light-induced instabilities in colloidal media**
  M. Matuszewski, W. Krolikowski, and Y. S. Kivshar, OE (2008).

- **Experimental Observation of Modulation Instability and Optical Spatial Soliton Arrays**
**Optical Forces**

In the Rayleigh regime (dipole approx.)

\[
p = 3V_p \varepsilon_0 n_b^2 \frac{m^2 - 1}{m^2 + 2} E_0
\]

\[
p = \alpha E_0
\]

\[
\therefore \alpha = 3V_p \varepsilon_0 n_b^2 \frac{m^2 - 1}{m^2 + 2}
\]

\[
m = \frac{n_p}{n_b}
\]

\[n_p > n_b \Rightarrow \alpha > 0\]

**Positive polarizability** (PP)

\[n_p < n_b \Rightarrow \alpha < 0\]

**Negative polarizability** (NP)
Optical Forces

Optical gradient force:

\[ \vec{F} = \frac{\alpha}{4} \nabla I \]

\[ \alpha = 3V_p \varepsilon_0 n_b^2 \left( \frac{m^2 - 1}{m^2 + 1} \right) \]

Positive polarizability (Attractive!)

\( n_p > n_b \Rightarrow \alpha > 0 \)

Negative polarizability (Repulsive!)

\( n_p < n_b \Rightarrow \alpha < 0 \)

The origin of the nonlinearity!

S. Stenholm, Rev. Mod. Phys. (1986)

Optical Radiation Pressure:

\[ \vec{F}_{\text{rad}} = \frac{\sigma}{c} \vec{S} \]

\[ \sigma_s = \frac{128}{3} \frac{\pi^5}{a^2} \frac{n_b^4}{\lambda} \left( \frac{a}{\lambda} \right)^4 \left( \frac{m^2 - 1}{m^2 + 2} \right)^2 \]
Optical Forces vs Polarizability

PP

Refractive index increases at beam center in both cases (artificial self-focusing)

NP

α > 0

α < 0
Engineering polarizability in colloidal suspensions?

- **Positive Polarizability (PP)**
  - Large scattering loss
  - Super-Kerr $\Rightarrow$ unstable

- **Negative Polarizability (NP)**
  - Self-cleaned channel
    - (Enhanced transparency)
  - Saturable $\Rightarrow$ stable!

- **Tunable nonlinearity**

**Mixed polarizability**
**Our experimental work:**

**synthetic nanosuspensions**

<table>
<thead>
<tr>
<th>Suspension</th>
<th>Particle Size</th>
<th>(n_p)</th>
<th>(n_b)</th>
<th>(n_p - n_b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP Polystyrene in Glycerin Water (3:1 ratio)</td>
<td>200nm</td>
<td>1.59</td>
<td>&gt;1.44</td>
<td>+0.15</td>
</tr>
<tr>
<td>NP PTFE in Glycerin Water (3:1 ratio)</td>
<td>200nm</td>
<td>1.35</td>
<td>&lt;1.44</td>
<td>-0.09</td>
</tr>
<tr>
<td>ZP PTFE in Glycerin Water (1:6 ratio)</td>
<td>200nm</td>
<td>1.35</td>
<td>=1.35</td>
<td>0</td>
</tr>
</tbody>
</table>

\(n_{\text{pure Glycerin}} = 1.47\)
\(n_{\text{water}} = 1.33\)
PTFE:Poly(tetrafluoroethylene) \(n_{\text{ptfe}} = 1.35\)

**First stable colloidal nanosuspensions with negative polarizibility!**
Beam propagation in tunable nanosuspensions

Experimental side-view

- $n_p > n_b$: Beam Collapse
- $n_p < n_b$: Enhanced transmission
- $n_p = n_b$: Diffraction

Thermal effect? No!

catastrophic self-focusing collapse severe scattering losses
Nonlinearity in Colloidal Suspensions

Nernst-Planck Eq:
(diluted sample)

\[ \vec{J} = \rho \vec{v} - D \nabla \rho \]

Drift due to gradient force

\[ \vec{v} = \mu \vec{F} \]

\[ \vec{F}_{\text{grad}} = \frac{\alpha}{4} \nabla I \]

Diffusion due to Brownian motion

At steady state condition:
\[ \vec{J} = 0 \]

\[ \rho(I) = \rho_0 \exp\left(\frac{\alpha}{4k_B T} I\right) \]

Maxwell-Garnett formula:

\[ n_{\text{eff}}^2 = n_b^2 \left(1 + 2f \frac{n_p - n_b}{n_b}\right)(I) = V_p \rho(I) \]

\[ \Delta n_{\text{NL}} = n_{\text{eff}}(I) - n_{\text{eff}}(I = 0) = (n_p - n_b)V_p \rho_0 \left(\frac{\alpha}{e^{4k_B T}} I - 1\right) \]

NLS –like Equation
- beam propagation in tunable nanosuspensions

Helmholtz eq: \[ \nabla^2 E + k_0^2 n_{\text{eff}}^2 E = 0 \]
\[ n_{\text{eff}} = (1 - f)n_b + fn_p \]

under SVEA: \[ E(x, y, z) = \varphi(x, y, z) \exp(ik_0n_bz) \]

\[ i \frac{\partial \varphi}{\partial z} + \frac{1}{2k_0n_b} \nabla^2 \varphi + k_0 \left( n_p - n_b \right) f \varphi + \frac{i}{2V_p} \sigma f \varphi = 0 \]

Exponential nonlinearity
Tunable

\[ \frac{\alpha I}{4k_B T} \approx \ln \frac{f}{f_0} + \frac{2B_2f_0}{V} \left( \frac{f}{f_0} - 1 \right) + \frac{3B_3f_0^2}{2V^2} \left( \frac{f}{f_0} \right)^2 - 1 \]

“Non-ideal gas” model
Virial coefficients \( B_{2,3} \)

Saturable for NP

ratio 0 - pure NP; 1 – pure PP
Optical Nonlinearities and Enhanced Light Transmission in Soft-Matter Systems with Tunable Polarizabilities

- Tunable nonlinearity by mixing PTFE and PS particles
- NP: Four-fold enhancement in transmission – “Self-induced transparency” (Fair comparison: starting at same initial linear transmission)
Interaction of self-trapped beams in NP suspensions

(out-of-phase repulsion)

Simulation (side-view propagation)

Experiment (transverse patterns)

Drive by optical forces - particle-density dependent;
Not mediated by thermal effects!
Such interaction not possible in PP suspensions.

Deep penetration achieved by forming dense shock fronts of particle concentration in PP (polystyrene) suspensions!

Advantages:

- much larger polarizability
- much deeper penetration (over 25 diffraction lengths)
- much lower power needed (mW)
- much more flexible in tuning optical response (through composition, size, and shape)...

Previous work: Metallic nanosuspensions

Closed aperture z-scan

$\text{CW laser beam}$

$\text{Sample 1 mm}$

$\text{Laser}$

$\text{CCD camera}$

Why so?

$n_2 < 0$

10mm

$n_2^{th}$ (cm$^2$/W)

$-1.0 \times 10^{-8}$

$-2.23 \times 10^{-8}$

$-2.92 \times 10^{-7}$

$-1.60 \times 10^{-8}$

$-1.941 \times 10^{-7}$

$-3.81 \times 10^{-8}$

Metallic nanosuspensions

$\sim 150$ mW

Due to thermal effects

Strong defocusing
## Samples used

<table>
<thead>
<tr>
<th># Sample</th>
<th>1 Gold nanorods</th>
<th>2 Silica-gold core-shells</th>
<th>3 Gold spheres</th>
<th>4 Silver spheres</th>
</tr>
</thead>
<tbody>
<tr>
<td>Parameters</td>
<td><img src="image1.png" alt="Gold nanorod" /></td>
<td><img src="image2.png" alt="Silica-gold core-shell" /></td>
<td><img src="image3.png" alt="Gold sphere" /></td>
<td><img src="image4.png" alt="Silver sphere" /></td>
</tr>
<tr>
<td>NP $\alpha_R &lt; 0$</td>
<td>100 nm</td>
<td>120 nm</td>
<td>40 nm</td>
<td>100 nm</td>
</tr>
<tr>
<td>NP $\alpha_R &lt; 0$</td>
<td>50 nm</td>
<td>15 nm</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

### Polarizability at 532 nm
- NP $\alpha_R < 0$
- PP $\alpha_R > 0$
NP sample: gold nano-rod

Polarizability

Extinction

\[ \alpha \left[ 10^{-32} \, \text{C} \, \text{m}^2 / \text{V} \right] \]

- \( \alpha_{\text{R}} \)
- \( \alpha_{\text{Im}} \)

- calculated
- measured

\( \sigma_e \) [norm.]

Wavelength \( \lambda \) [nm]
NP samples: Gold nano-rod suspension:

**Polarizability**

\[ a \left( \times 10^{32} \text{ C}^2 \text{ m}^{-3} \text{ V}^{-1} \right) \]

\[ a \]

\[ a_l \]

\[ a_r \]

**linear:**

10mW

**nonlinear:**

250mW

10mW

100mW

200mW

250mW

3 mm

1 mm

0.3 mm

0.1 mm
NP sample: Silica core-gold shell suspension

Polarizability tuning
Core-shell nanosuspension:

Fivefold larger NP

Nano-rod suspension (direct comparison)
PP Samples: gold & silver spheres:

\[ i \frac{\partial \phi}{\partial z} + \frac{1}{2k_0 n_b} \nabla^2 \phi + k_0 \left( n_p - n_b \right) \rho V \phi - k_0 \Delta n_T \left| \phi \right| + \frac{i \sigma \rho}{\varepsilon} \phi = 0 \]

\[ \Delta n_T = \left( \frac{\partial n_b}{\partial T} \right) (T - T_b) (1 - f_V) \]

Beam does not collapse as in dielectric PP suspensions!
Guiding light by light in metallic nanosuspensions:

**Pump (soliton) beam:** $\lambda = 532\text{nm}, P=40\text{mW}$

**Probe (guided) beam:** $\lambda = 1064\text{nm}, P=50\text{mW}$

(probe beam itself has no (or weak) NL self-action!)

Controlling strong IR beams by a weak green beam!

*SPR Peak: 527 nm*

**Guided:** FWHM 150 $\mu$m

**Unguided:** FWHM 670 $\mu$m
Outlook

• Dielectric, metallic, and biological colloidal suspensions are fun to play with;
• Nonlinear optical manipulation in such highly scattering suspensions is still unexplored;
• New opportunities open up in developing soft-matter systems with engineered nonlinearities.
Thank you for your attention!

Website: http://www.physics.sfsu.edu/~laser/

Co-worker:
Shima Fardad and D.N. Christodoulides - CREOL
Weining Man, Z. Zhang, Anna Bezryadina - SFSU