Enhancing and controlling light with plasmonic and non-plasmonic nanoantennas

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Presented by Pablo Albella
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Outline

Part I:
• Surface Enhanced Spectroscopies (SES)
  • Introduction to plasmonic nanoantennas and its limitations

Part II:
• Non-plasmonic HRI dielectric nanoantennas as an alternative to plasmonics for enhancing light (in near and far field) with low losses
  - Electric and Magnetic Hot Spots
• Experimental demonstration of these novel type of nanoantennas able to enhance Raman and Fluorescence:
  - Heat assessment based on thermometry
Outline

Part III:

- What about enhancing light with directional control? or switchable steering of light?
  - Possible direct application in Selective sensing or SES
  - Application in optical nano-circuitry

- Polarization control of high transmission / reflection switching by all-dielectric metasurfaces

Part IV:

- Dielectric - Metal hybrid structures for efficient THG

Part V:

- Examples of (bio) - sensors
  - MO-Kerr effect in magnetoplasmonic crystals
  - DNA mapping with nanopipettes (SERS,SEF)
Plasmonic antennas are widely studied due to their capability to convert free propagating radiation into highly enhanced-localized fields.

Part I: Introduction to SES and plasmonic nanoantennas

- **Idea in SES:** Using the enhanced electromagnetic near and far field of a nanostructure oscillating in resonance

- Pre-condition for SES[#, *]: Matching molecular vibration of interest and resonant excitation of the nanoantenna

- Very important to tune the resonance precisely! And with strong near and Far field enhancement!!

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  - Distinct Spectral Shifts and strong local field enhancements (broadly applied in SES)

Neubrech, Aizpurua, et.al, PRL 101, 157403 (2008)

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Near-IR photodetector
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  - Scattering will follow the 4th power SERS law
  - Widely accepted, but not clearly shown why, until 2012*

Part I: Introduction to SES and plasmonic nanoantennas

- **Idea:** Using the enhanced electromagnetic near and far field of a nanostructure oscillating in resonance

  - All that is possible, because the Scattered intensity $I$ scales with the 4th power of the local field enhancement at the metal surface.

  - Direct and Quantitative verification of this law and its **underlying electromagnetic scattering mechanism was more than a challenge to tackle.**

  - Why? Difficulties arise from the complex processes typically involved in surface-enhanced spectroscopies:
    - Chemical bonding
    - Charge transfer between object and metal nanostructures

- **How can we isolate the electromagnetic mechanism?**

Surface Enhanced Spectroscopy
Schematics of a surface-enhanced light scattering Process

Schematics of typical Antenna-enhanced Raman process (SERS)

Elastic antenna-enhanced scattering process

Inelastic scattering process from an object (O) in the presence of a metal nanostructure

We rely on the underlying electromagnetic mechanism of the signal enhancement being challenging to trace experimentally.
Surface Enhanced Spectroscopy

Schematics of a surface-enhanced light scattering process

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Elastic antenna-enhanced scattering process

Inelastic scattering process from an object (O) in the presence of a metal nanostructure

By relying on elastic scattering (s-SNOM) we were able to isolate the electromagnetic effect!!

IR Spectroscopy of plasmonic antennas with S-SNOM. How?

Spectroscopy in frequency is equivalent to spectroscopy in length.

IR Light $\lambda = 11.1 \, \mu m$ fixed
AIM: To relate the scattered field $E_n$ with the local field enhancement $f$ and proof that $E_n = f^2 \Rightarrow I_n = f^4$

1. We measure $|E_n|$ and $\Delta \phi_n$ at the hot spot (x)

2. We compare them to the numerically calculated $|f|$ and $\Delta \phi_f$ of the local field enhancement.

We clearly see the resonance behaviour, with the resonance appearing at $L = 3.7 \, \mu m$. 

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s-SNOM spectra do not agree with calculated near-field spectra!!

Single Rod

- Calculated near-field enhancement $f$
  - Measured s-SNOM amplitude

- Calculated near-field phase $\phi$
  - Measured s-SNOM phase

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Square of the near field agrees well with s-SNOM data

**Single Rod**

- Calculated near-field enhancement $f$
- Measured s-SNOM amplitude
- Squared calc. near-field enhancement $f^2$

---

Parametric representation (Intensity and Phase)

Single Rod

Intensity

\[ \log[I_n(L)] = 4.08 \cdot \log[f(L)] \]

Phase Shift

\[ \Delta \phi_n = 1.85 \cdot \Delta \phi_f \]

How we can understand this observation?

\[ E_{\text{loc}} = f E_{\text{inc}} \]

**Elastic** antenna-enhanced scattering process

Antenna-enhanced Raman (inelastic) process (SERS)

\[ E_{\text{inc}}(\omega) \quad d(\Omega) \]

\[ E_{\text{inc}}(\omega_1) \quad E_{\text{loc}} \]

How we can understand this observation?

\[ E_{\text{loc}} = f E_{\text{inc}} \]
\[ E_{\text{AOA}} \propto f^2 E_{\text{inc}} \]

\[ f = \left| f \right| e^{i\varphi} \]

Field enhancement \( f \) is a complex value!

\[ \left| E_{\text{AOA}} \right| \propto f^2 \Rightarrow I_{\text{AOA}} \propto f^4 \]
\[ \varphi_{\text{AOA}} = 2\varphi \]

Elastic antenna-enhanced scattering process

Antenna-enhanced Raman (inelastic) process (SERS)

Double role of the antenna: illuminating the object and scattering off the object.

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How we can understand this observation? Other view...

1. \( \varepsilon_{np} \)

2. \( E_{inc}(\lambda_{inc}) \), LSP

3. NP, Target

4. Target Scattering \( \propto E_{inc} \)

5. Reradiation process

\[ E_R \propto f(\lambda_T) \cdot f(\lambda_{inc}) \cdot E_{inc} \]

\[ I_R \propto f^2(\lambda_T) \cdot f^2(\lambda_{inc}) \cdot I_{inc} \]
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- All these is well known!

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- **Good results in SERS, SEIRA and SEF applications, have been reported over the last 20 years...**

- All these is well known! But what about losses and Heating issues in the nanoantennas?

- **Can dielectric nanoantennas play a new role in SES?...**

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Influence of losses/heat radiation to the efficiency of Surface Enhance Spectroscopies

• An effect of the losses in plasmonic structures, and an aspect quite often neglected with respect to SES, is the **local heating** of the particle due to the absorption of incident radiation and the transduction into thermal energy.

• Theoretical and experimental studies have reported temperature increases ranging from 50 K under continuous excitation to as high as 1000 K using pulsed light sources [#].

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- Theoretical and experimental studies have reported temperature increases ranging from 50 K under continuous excitation to as high as 1000 K using pulsed light sources.

- This increase in the particle $T$, and in turn in the $T$ of the surrounding medium, can directly influence the SERS and SEF signals.

  - This fact has been attributed to several processes (thermal annealing, modified adsorption/desorption kinetics of surface molecules, and changes in the dielectric properties of the medium and NP) [*].

Plasmonic nanoantennas show strong hot spots but...

- **Metals (plasmonics) can offer very high relative field enhancements**
  - However, they exhibit high Ohmic losses that lead to local heating
    - Fluorescence quenching without spacer layers [*]
    - Can cause localised damage to the sample or molecules under study [**]

- Au NPs functionalised with DNA chains acting as spacer between fluorophore and nanoantenna.
  - SEF significant for distances larger than 10nm

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- This would also damp the Raman strongly. Novotny quantitatively measured the continuous transition from fluorescence enhancement to fluorescence quenching on a single molecule.

Alternatives to address this problem

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**The question is:** Can we get around these issues? Is it possible to achieve enhanced NF and FF but under a low loss/heat response?

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The question is: Can we get around these issues? Is it possible to achieve enhanced NF and FF but under a low loss/heat response?

The answer is: Yes, Non-Plasmonic Nanoantennas can!
  - Nanostructures made of dielectrics with high refractive index - Si, Ge, GaAs, GaP
  - Important to note that we refer to individual structures, not metasurfaces!

A bit of literature review on HRI dielectrics...

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- Theoretically explored the concept of coupling dielectric antennas with electric or magnetic dipolar emitter.
- Experimentally demonstrated those findings either in the visible, near IR or microwave...

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Our aim is: Non-Plasmonic (all-dielectric) Nanoantennas for SES

Theoretical Proposals

First Experimental Proof

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J. J. Saenz et al, JOSA A
Y. Kivshar et al. (Hybrid) ACS Nano
F. Moreno et al. Nat. Comms.
J. Aizpurua et al. Opt. Express
2013 - B. Luk’yanchuk et al. Nat. Comms
P. Albella et al. Jphys Chem C
P. Albella et al. ACS Photonics
2014 - P. Albella et al. ACS Photonics
B. N. Chichkov et al. Nat. Comms
A. I. Kuznetsov et. al. Nano Letters
Y. Kivshar et. al. ACS Nano
2015 - G. Yang et. al. ACS Nano
G. Yang et. al. Nat. Comms
S. Maier et al. Nat. Comms

Pablo Albella @ PHOTOPTICS 2018 (27 JAN 2018) - Madeira, Portugal
Non-Plasmonic HRI Nanoantennas

Scattering of Si particle ($r = 230$ nm)

- Strong magnetic resonances
- Field enhancement inside particle

*A. Garcia-Etxarri et al., Opt. Express. 19, 4815 (2011)*
In 2013 we theoretically showed how Silicon dimers offer not only, near and far field electric enhancement but also magnetic, both under very low-losses [#].

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The Near and Far Field Electric and Magnetic Field enhancements are due to the interaction between electric and magnetic Mie modes, and can be spectrally tuned.

\[
\begin{align*}
E_{\text{cat}}(r) &= \sum_{j=1,2} \left\{ \frac{k^2}{\epsilon_0 \epsilon_h} \mathcal{G}_E (r - r_j) \cdot \mathbf{p}_j + iZk^2 \mathcal{G}_M (r - r_j) \cdot \mathbf{m}_j \right\} \\
H_{\text{cat}}(r) &= \sum_{j=1,2} \left\{ -i \frac{k^2}{Z \epsilon_0 \epsilon_h} \mathcal{G}_M (r - r_j) \cdot \mathbf{p}_j + k^2 \mathcal{G}_E (r - r_j) \cdot \mathbf{m}_j \right\}
\end{align*}
\]

HRI Nanoantennas in the VIS: Hot Spots and FF Enhancement

Near Field Response

- Au dimers offer better NF enhancement when compared with the Non-plasmonic ones.

*P. Albella, R. Alcaraz de la Osa, F. Moreno and S. A. Maier. **ACS Photonics** 1, 524-529 (2014)
HRI Nanoantennas in the **VIS**: Hot Spots and FF Enhancement

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HRI Nanoantennas in the VIS: Hot Spots and FF Enhancement

Near Field Response

• Au dimers offer better NF enhancement when compared with the Non-plasmonic ones.

Far Field Response

• HRI antennas show larger FF enhancement.
• More Scattering efficiency together with the possibility of increasing the incident power can compensate the smaller NF enhancement in SES applications

*P. Albella, R. Alcaraz de la Osa, F. Moreno and S. A. Maier. ACS Photonics 1, 524-529 (2014)
Theoretical Proposal
Non-Plasmonic nanoantenas: low local temperature

- We have seen that HRI dielectric nanoantennas can generate hot spots and good scattering efficiency.
- The idea of looking for alternatives to plasmonic nanoantennas came up from the large absorption and therefore heat that they generate in their surrounding.
Theoretical Proposal
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• The idea of looking for alternatives to plasmonic nanoantennas came up from the large absorption and therefore heat that they generate in their surrounding.

• Metals can produce extreme changes in local temperature (implications for SERS and SEF)
  • Damage to antennas and molecules
  • Can modify the local environment (n) and/or possibly degrade the performance
Experimental Demonstration:
Low-Heat SERS and SEF with Si-Dimers

- Resonance is a mix between electric and magnetic modes in Si (purely electric in Au)
- Reasonable NF enhancement for Si, although not as high as Au (but FF enhancement and hot spot volume compensates this).

Si Dimers D=220nm, h=200nm, gap~20nm fabricated with reactive ion etching on Si-on-insulator wafers

- Resonance is a mix between electric and magnetic modes in Si (purely electric in Au)
- Reasonable NF enhancement for Si, although not as high as Au (but FF enhancement and hot spot volume compensates this).
- Higher E/E0 can be achieved by engineering future dielectric nanoantenna configurations

Experimental proof of SERS and SEF with the same Si-Dimers

- Sample shows both enhanced Raman scattering and enhanced fluorescence

PMMA molecules

Nile Red molecules
Assessment of Local Heat based on Molecular thermometry

- Local heat generation around the NA was determined using molecular thermometry.
- 2 lasers:
  - Heating laser on resonance with the NA.
  - Imaging laser
- Spectral shifts and reduction in intensity of nile red molecules upon increase in T.

Heating laser $\lambda = 860-890$nm
Low power Imaging laser $\lambda = 532$nm

Reference: fluorescence images without the heating laser turned on.
- Si produce enhanced fluorescence larger than Au

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Low power Imaging laser $\lambda=532$nm

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  • Si produce enhanced fluorescence larger than Au

Fluorescence images with the heating laser on at 6 mW $\mu$m$^{-2}$
  • Intensity clear drops for the Au antennas.

Local Heat Mapping results: Gold vs Si Nanoantennas

- Si antennas show little heating while substantial heating is observed for Au antennas.
- Modelling the heating and extracting Tgap indicates local temperatures of > 100 °C at high powers.
- Very good agreement with theory (dotted lines).
Summary of Part II

• We have shown a novel type of nanoantenna based on all-dielectric materials (Si in this case)

• These nanoantennas enhanced the Raman scattering of a polymer thin film by a factor of \( \sim 10^3 \) and also allowed surface enhanced fluorescence by a factor of \( \sim 2 \times 10^3 \)
  - avoiding the well-known fluorescence quenching effects observed for metallic structures when no spacer layers are used.

• Molecular thermometry demonstrate that dielectric nanoantennas produce ultra-low heating, thus overcoming one of the main drawbacks of traditional plasmonic materials.
  - important advances in many fields can be foreseen due to almost no-restrictions in the power that can be delivered to these non-plasmonic nanoscale devices
  - Some examples: nanoelectronics or unperturbed sensing of nanoemitters behaviour by SES.
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Introduction to Unidirectional forward scattering (Kerker’s first condition)

- When the first Mie coefficients, $a_1$ and $b_1$, corresponding to the electric and magnetic dipolar resonances are equal, the backward scattering is suppressed (1$^{st}$ Kerker condition)
  - Unidirectional forward scattering is achieved due to the interference between the electric and magnetic resonances

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First demonstrated in the microwave (radiation patterns measured in Fresnel Institute (anechoic chamber))

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- [A. Gracia-Etxarri et al., Opt. Express. 19, 4815 (2011)](http://opticalengineering.org/content/19/17/4815)

Low scattering efficiency
(electric and magnetic resonance far from each other)

Some attempts to increase the scattering efficiency...

*Spheroids and low aspect ratio disks can increase the efficiency*

Boris S. Luk’yanchuk *et al.* ACS Photonics 2015, 2, 993–999

I. Staude *et al.* ACS Nano 2013, 7, 7824–7832

Measured optical transmittance and reflectance spectra for Si nanodisks embedded into a low-index medium.
F/B ratio based on optimized single disk with low aspect ratio

I. Staude et al. ACS Nano 2013, 7, 7824–7832

Optimised Single disk (Excited by PLANE wave instead)
Asymmetric dimer configuration to enhance and direct light

- Idea: combine HRI dielectric nanoparticles of different sizes.
  - the position and intensity of the electric and magnetic resonance of dielectric particles strongly depend on their sizes.
  - Tuning them to achieve overlapping of those resonances could open up new possibilities for novel optical properties.

![Diagram of scattering cross-section vs. wavelength (S1 in red, S2 in blue)]
Optimizing dimer size to achieve first kerker condition

Asymmetric dimer

Silicon Dimer: $D_1 = 165$ nm, $D_2 = 225$ nm
Gap = 20nm

Overlap of electric and magnetic resonance

Scattering spectra of single spheres

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Verification of the first Kerker condition

- Electric and magnetic dipoles were calculated considering the electric-electric, magnetic-magnetic and electric-magnetic dipole couplings.
  - Based on the Analytical dipole-dipole model proposed in [*]

\[ R_e = \Re \left( \frac{1}{\varepsilon_0 \varepsilon} \left( p_{1y} + p_{2y} \right) \right) \]
\[ R_m = -\Re \left( Z \left( m_{1x} + m_{2x} \right) \right) \]
\[ I_e = \Im \left( \frac{1}{\varepsilon_0 \varepsilon} \left( p_{1y} + p_{2y} \right) \right) \]
\[ I_m = -\Im \left( Z \left( m_{1x} + m_{2x} \right) \right) \]

\[ R_e = R_m \]
\[ I_e = I_m \]

at \( \lambda = 650 \text{ nm} \), verifying the achievement of the first Kerker condition.

F/B ratio of the asymmetric dimer: comparison to a single disk in low aspect ratio

- DF Scattering CS Calculation (Collection of scattered field on the forward and backward hemisphere)

Spherical asymmetric dimer
(20nm gap)

Basic Principle: No substrate

Forward / backward ratio comparable between the asymmetric dimer and optimised single disk.

* I. Staude, A. E. et.al, ACS Nano, 2013, 7, 7824–7832

# T. Shibanuma, P. Albella and S. A. Maier. Nanoscale, 2016, 8, 14184
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# T. Shibanuma, P. Albella and S. A. Maie. Nanoscale, 2016, 8, 14184
High F/B ratio of the asymmetric dimer and Presence of Hot Spot

Using asymmetric silicon dimer

Electric field mapping at $\lambda = 647 \text{ nm}$

- This hot spot can not be obtained in single disk or spheroid cases.
- Unidirectional forward scattering with high intensity can be achieved
- It can be used as a novel nanoantenna for directional SERS or SEF

# T. Shibanuma, P. Albella and S. A. Maier, Nanoscale 2016, 8, 14184
Experimental demonstration

- Dark field spectroscopy for measurement of scattering from single nanoantenna
- This setup enables us to measure only scattering, excluding incidence.

Incidence: $\theta_i = 60-70$ $^\circ$
Scattering: $\theta_s = 0-53$ $^\circ$

- Fabricated sample and forward / backward scattering

Peak and valley observed in forward and backward scattering, respectively.
→ Unidirectional forward scattering with high efficiency

# T. Shibanuma, P. Albella and S. A. Maier, *Nanoscale* 2016, 8, 14184
Experimental demonstration: Comparison with theory and influence of incidence angle

As incident angle decreased, the F/B ratio increased and reached around 15 with nearly normal incidence.

# T. Shibanuma, P. Albella and S. A. Maier, *Nanoscale* 2016, 8, 14184

Presented by Pablo Albella
Extending the idea for multi-wavelength and broadband response

Three Si spheres aligned with diameter of 165 nm, 225 nm, and 310 nm
When dielectric nanoantennas are aligned periodically, they can act as a metasurface providing with an effective permittivity.

When the two peaks overlap,
\[ \varepsilon \sim \mu \]
\[ \Rightarrow \quad \text{High transmission} \]

When the two peaks separate,
\[ \varepsilon \text{ and } \mu \text{ have opposite signs} \]
\[ \Rightarrow \quad \text{High reflection} \]

(I. Staude et al., ACS Nano, 7, 7824 (2013)

(\varepsilon, \mu: \text{effective permittivity and permeability of dielectric metasurface})
Polarization control of high transmission / reflection switching by all-dielectric metasurfaces

• Basic idea
  - **Spectral overlap** of electric and magnetic dipoles \(\rightarrow\) **High transmission**
  - **Spectral separation** of electric and magnetic dipoles \(\rightarrow\) **High reflectance**
Polarization control of high transmission / reflection switching by all-dielectric metasurfaces

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• Array of the Si dimer
  - **Polarization along the dimer axis** → Mode hybridization induces spectral overlapping

Scattering spectra of Si sphere dimer
Diameter: 300 nm, Gap: 10 nm
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  - **Spectral overlap** of electric and magnetic dipoles  $\rightarrow$ **High transmission**
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- Array of the Si dimer
  - **Polarization perpendicular to dimer axis**  $\rightarrow$ Mode hybridization induces spectral separation

Scattering spectra of Si sphere dimer
Diameter: 300 nm, Gap: 10 nm
Proof of concept: array of nanospherical dimers

Si spherical dimer in air

FDTD calculation

High R <-> High T switching around $\lambda = 975$ nm

Experimental demonstration: array of nanocylindrical dimers

Array of cylindrical Si dimer on sapphire substrate

FDTD calculation

Experiment (FTIR)

At $\lambda = 1688$ nm
TM: 99% transmittance
TE: 95% reflectance

At $\lambda = 1718$ nm
TM: 86% transmittance
TE: 77% reflectance

What about: switchable and tunable steering of light WITH Low Loss

- **Experimental demonstration:** ACS Photonics 4, 489-494 (2017)

What about light steering?

*Sensing and Selective SES*
Tuneable directional scattering either to left or right direction

• Next topic is about directional control of scattering in the left or right direction from the incident axis

Bimetallic configuration

V shape nanoantenna


• Most Scattering into the substrate.

• Control of the scattering light along the substrate would be preferable for some applications (e.g. optical nanocircuitry)
Basic idea to switch in-plane scattering either left or right

Using asymmetric dimer

- Drastic phase shift by dimensions
- Tuneable from visible to microwave

Dimer of Silicon Spheres:
\( D_1 = 150 \text{ nm}, \ D_2 = 230 \text{ nm}, \ d = 8\text{nm} \)

Extinction Calculated by the dipole-dipole model [*]

Basic idea to switch in-plane scattering either left or right

- Scattering direction defined as the angle where maximum scattering is achieved.
- Direction can be changed depending on the wavelength of excitation.

- Light scattered to either left (+18°) or right (-52°) depending on the wavelength.

Asymmetric silicon dimer for tuneable scattering along the substrate

Si dimer on a silica substrate

Far field pattern in the xy plane calculated by FDTD

Interference between the excited magnetic dipoles results in the tuneable scattering

To experimentally demonstrate the tunable directional scattering along the substrate, a BFP (Back focal plane imaging) technique combined with a prism coupling was designed.

- Basically, an evanescent wave is generated by total internal reflection and travels along the substrate until it excites the nanoantenna.
- The scattering lobe is then observed with high NA objective.

Experimental results vs numerical predictions

Scattering direction can be tuned by the excitation wavelength

This idea inspired recent works (optical computing)

**What a Twist: Silicon Nanoantennas Turn Light Around**

Published: November 16, 2016.
Released by Moscow Institute of Physics and Technology.

Nanoantenna Changes Direction of Light and the Prospects of Optical Computing

Belov and Alu, Laser Photonics Rev. 10, No. 6, 1009 (2016)

A team of physicists from ITMO University, MIPT, and The University of Texas at Austin have developed an unconventional nanoantenna that scatters light in a particular direction depending on the intensity of incident radiation. The research findings will help with the development of flexible optical information processing in telecommunication systems.

Scattering cross-section of isolated nanoparticles with increasing Electron Hole pair density in the resonant particle.
Part III:
- What about enhancing light with directional control? or switchable steering of light?
  - Possible direct application in Selective sensing or SES
  - Application in optical nano-circuitry

- Polarization control of high transmission / reflection switching by all-dielectric metasurfaces

Part IV:
- Dielectric - Metal hybrid structures for efficient THG

Part V:
- Examples of (bio) - sensors
  - MO-Kerr effect in magnetoplasmatic crystals
  - DNA mapping with nanopipetes (SERS,SEF)
Introduction to THG

• Third-order susceptibilities of metals are among the highest in nature
  - enables, in principle, excellent THG performances.

• Skin depth of conductors is generally small
  - any third-order nonlinear effect from metals results in strongly reduced efficiencies.

• To further enhance the THG process, plasmonic nanostructures have been combined with non-metallic nm-scaled nonlinear materials.
  - For example, by placing an ITO nanoparticle at the hot spot of a metallic nanodimer.
Metal – dielectric hybrid structures for efficient THG

- Third harmonic generation (THG) from high ref index dielectric nanostructures

- Efficiency is still low, because of the low electric field enhancement in dielectric structures

\[ \eta_{TH} \approx 0.0002\% \]

Metal – dielectric hybrid structures for efficient THG

- Third harmonic generation (THG) from high ref index dielectric nanostructures

- Efficiency is still low, because of the low electric field enhancement in dielectric structures

- Ge has shown conversion efficiencies of ~0.001% at 550 nm (green)
  - Performance cannot be extended to the blue region of the visible spectrum, due to high absorption of Ge below 1600 nm

\[ \eta_{TH} \approx 0.0002\% \]

\[ \eta_{TH} \approx 0.001\% \]


How to improve the TH conversion efficiency?

In Third order nonlinear phenomena, the density of third harmonic dipoles are basically proportional to the third power of electric field (THG intensity is proportional to the 6th power of E).

The idea is combining a metallic ring with a dielectric nanostructurecore to make hybrid one.
- As we showed in [*] Plasmonic resonance of a Au nanoring can enhance the E field in a relatively large volume.

This can boost the anapole mode supported by a Si nanodisk
- strongly enhancing the electric field inside the large third-order susceptibility dielectric

[*] A. Rakovich, P. Albella and S. A. Maier. ACS Nano, 9, 2648 (2015)
Excitation of anapole mode for intense electric field

- To predict THG capabilities of the hybrid structure
  - we need to evaluate its ability to **concentrate the electric field inside the dielectric core**
  - by measuring and calculating first the extinction (a clear valley observed around 1325 nm)
Excitation of anapole mode for intense electric field

- To predict THG capabilities of the hybrid structure
  - we need to evaluate its ability to **concentrate the electric field inside the dielectric core**
  - by measuring and calculating first the extinction (a clear valley observed around 1325 nm)

- we numerically explore the normalized electric field intensity ($|E|^2/|E_0|^2$) averaged within the Si nanodisk volume, using the following expression:

  $$ F = \frac{\iiint |E|^2 \, dV}{|E_0|^2 V} $$

  - at the wavelength of the extinction valley, drastically increased.
Let’s double check the excitation of anapole mode and the increase in $F$.

Au nanoring can enhance the electric field inside.

[Graph showing electric field distribution]
Let’s double check the excitation of anapole mode and the increase in $F$

Au nanoring can enhance the electric field inside.

Comparing the isolated structures with the hybrid structure shows $E/E_0 \sim 9$.

The shape of the distribution is quite unique, (anapole mode). Its physical origin has been theoretically attributed [*] to the destructive interference in the far field between the radiation patterns produced by the electric and toroidal dipole modes.

This provides a route to maximize the electric field energy inside the particle

• The **hybrid structure shows very strong TH intensity**, 1000 times higher than single Si disks, and 100000 times higher than the Au ring

The hybrid structure shows very strong TH intensity, 1000 times higher than single Si disks, and 100000 times higher than the Au ring.

By sweeping the pumping power, we found the slope of 3 in the logarithm scale of TH intensity, confirming that this is third harmonic generation.

- The TH conversion efficiency of the hybrid structure: $\eta_{TH} \sim 0.007 \%$
- This is as far as we know the largest TH efficiency from a nanoantenna.

Very important: scalability offers THG throughout entire VIS

THG enhancement can be achieved throughout the visible regime

Summary of Part IV

• Hybrid structures can generate strong TH emission in the optical range.

• The anapole mode supported by the dielectric core, boosted by the plasmonic resonance of the surrounding metal nanoparticle, produced high electric field enhancement within the Si nanostructure.

• TH conversion efficiency can be drastically improved due to the coupling of the individual components defining the hybrid, (up to 0.007%).

• The optimum emission wavelength can be tuned from the blue to the red region of the visible spectrum by suitably adapting the nanosystem geometrical dimensions.
Outline

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Part IV:
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Magnetoplasmonic Crystals for the Design of Highly Sensitive Plasmonic (Bio)sensing Platforms

- An example of a simple yet highly sensitive sensing platform can be made as a one-dimensional magneto plasmonic crystal [1]

- Sensitivity of an SPR-based sensor depends on where the EM field is most amplified
  - IDEA: develop a magnetoplasmonic crystal to excite SPRs mainly localized at the analyte region.
    - This is done optimizing the geometry of the grating and the MO metallic slab at the $\lambda_{\text{inc}}$ [2].

- By using the optimization procedure in [3], enhanced TMOKE values with very narrow Fano-like resonant peaks can be achieved

- These Fano-like resonances are extremely sensitive to the refractive index of the surrounding media, thus allowing to detect very small changes in the dielectric properties of the analyte.

Another example: DNA mapping with nanopipette (SERS and TERS)

Gold-Glass nanopipette

On-Demand Surface- and Tip-Enhanced Raman Spectroscopy Using Dielectrophoretic Trapping and Nanopore Sensing

Kevin J. Freedman, †,⊥ Colin R. Crick, †,⊥ Pablo Albella, † Avijit Barik, §,‖ Aleksandar P. Ivanov, †
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Supporting Information

ABSTRACT: Surface-enhanced Raman spectroscopy (SERS) and tip-enhanced Raman spectroscopy (TERS) have shown great promise in the detection and analysis of trace analytes throughout numerous fields of study. Both SERS and TERS utilize nanoscale plasmonic surface features to increase the intensity of observed Raman signals by many orders of magnitude (> 10^6). One of the major factors limiting the wider and more routine implementation of the enhanced Raman phenomena is in the difficulty of forming consistent and reliable plasmonic substrates with well-defined "hot-spots". We address this limitation by designing a platform that can be used for both SERS and TERS. The presented technique allows for rapid, controlled, "on-demand", and reversible formation of a SERS substrate using dielectrophoresis at the end of a nanoscale pipet. This drives gold nanoparticles in solution to concentrate and self-assemble at the tip of the pipet, where analytes can be detected effectively using SERS. An additional benefit of the platform is that the nanopipet containing a nanopore can be used for detection of individual nanoparticles facilitated by the added enhancement originating from the nanopipet tip enhanced signal. Complementing the experimental results are simulations highlighting the mechanism for SERS substrate formation and TERS detection.
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