Dielectric-based nanoantennas for surface enhanced spectroscopies and nonlinear photonics

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Si vs. Au nanoantennas for SERS

GaP nanoantennas for SEF and SHG

THG and FWM from Ge nanodisks

THG from hybrid Si/Au nanoantennas

Conclusions

Publications:
- G. Grinblat et al. ACS Photonics, 10.1021/acsphotonics.7b00631 (2017)
Si nanoantennas vs. Au nanoantennas - SERS


Scattering and absorption - Au

Scattering and absorption - Si

SERS factor - Au

SERS factor - Si
Si nanoantennas vs. Au nanoantennas - heating

By increasing ~5 times the power at $\lambda=860$ nm for the Si nanoantenna, it is possible to obtain the same Raman signal as in the case of Au, but producing 75% less heating.

GaP nanoantennas – Scattering cross section

Absorption coefficient

\[ \text{Abs. coeff. (1/cm)} \]

\[ \lambda \text{ (nm)} \]

\[ \text{Au} \]

\[ \text{Si} \]

\[ \text{GaP} \]

Schematic of the experiment

\[ E_{\text{fluo}} \]

\[ E_{\text{exc}} \]

SEM image

Scattering cross section

Field intensity distribution (X-pol)

Field intensity distribution (Y-pol)

GaP nanoantennas – SEF


Fluorescence spectra

Fluorescence emission decay

Decay times

<table>
<thead>
<tr>
<th></th>
<th>(\tau_0) (ns)</th>
<th>(\tau_1) (ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Backgr.</td>
<td>1.1</td>
<td>-</td>
</tr>
<tr>
<td>X-Pol</td>
<td>1.1</td>
<td>0.12</td>
</tr>
<tr>
<td>Y-Pol</td>
<td>1.1</td>
<td>&lt;0.05</td>
</tr>
</tbody>
</table>

\[
I = A_0 e^{-t/\tau_0} + A_1 e^{-t/\tau_1}
\]

Effective excitation volume

Fluorescence enhancement factors

\[
F_{X,Y} = \frac{V_{\text{spot}}}{V_{\text{eff}(X,Y)}} \frac{I_{X,Y}}{I_{\text{ref}}}
\]

Fluorescence enhancement

X-pol 3600

Y-pol 155
GaP nanoantennas – SHG

SEM image and schematic of the experiment

Scattering cross section

Electric field intensity

Nanodisk vs. Bulk - SHG

\[ \eta_{\text{SHG}} = 0.0002\% \]

The SHG intensity provides a good measure of the square of the surface electric energy. The SHG intensity for nanodisks of different radii is shown alongside the graph of the square of the surface electric energy as a function of wavelength. The equation for the square of the surface electric energy is given as:

\[ W_E^{(S)} = n^2 \int |E|^2 dS/2 \]
Ge nanoantennas - THG

**Refractive index**

![Graph showing refractive index vs. wavelength for different materials, including Ge, Si, As₂Se₃, Sl₃N₄, and SiO₂.]

**Nonlinear index**

![Graph showing nonlinear index vs. wavelength for different materials, including Ge, Si, As₂S₃, Sl₃N₄, and SiO₂.]

**Large field enhancement**

![Image of large field enhancement with an antenna and light field illustration.]

**Large third-order susceptibility**

![Image illustrating large third-order susceptibility with Eₜ and Eₜ.]

Nanophotonics 2014; 3(4-5): 247–268

Faraday Discuss., 2015, 178, 185

Nano Lett. 2014, 14, 6488–6492
Ge nanodisks – Extinction and electric energy


Experimental extinction

Simulated extinction

Electric energy

\[ W_E = \frac{n^2}{2} \int |E|^2 dV \]

R_1 = 350 nm
R_2 = 635 nm
R_3 = 700 nm

SEM image

Electric field intensity \(|\frac{|E|^2}{|E_0|^2}|\)
Ge nanodisks – THG

The TH intensity is a good measure of the cube of the inner electric energy

Maximum efficiency of 0.001%!

Integrated THG intensity

Simulation – (Electric energy)$^3$

Nonlinear performance

Integrated TH intensity (a.u.)

TH wavelength (nm)

Integrated TH intensity (a.u.)

Fundamental wavelength (nm)

Simulation – (Electric energy)$^3$

Nonlinear performance

Fundamental wavelength (nm)

Reference values for THG efficiency

<table>
<thead>
<tr>
<th>Reference</th>
<th>Method</th>
<th>Efficiency ($10^{-4}$%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nano Lett. 14, 6488 (2014)</td>
<td>Si nanodisk–Dipolar magnetic mode</td>
<td>0.1</td>
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<tr>
<td>Nano Lett. 15, 7388 (2015)</td>
<td>Si metasurface–Fano resonance</td>
<td>1</td>
</tr>
<tr>
<td>Nano Lett. 16, 4857 (2016)</td>
<td>Si quadrumer–Fano resonance</td>
<td>1</td>
</tr>
<tr>
<td>Nanoscale 9, 2201 (2017)</td>
<td>Si dimer</td>
<td>2</td>
</tr>
</tbody>
</table>
Increasing disk radius

SEM/THG/ \(|E|^2/|E_0|^2\) for different disk sizes

Third harmonic is generated in enhanced field regions.

We can map the electric field distribution!

**Ge nanodisks – FWM**

*G. Grinblat et al. ACS Photonics, 10.1021/acsspectrosc.7b00631 (2017)*

**Schematic of the experiment**

Nonlinear cross sections between HOAM and HOM

**Nonlinear optical response of the nanodisk**
Si/Au hybrid – Extinction, E-field, electric energy


**Absorption coefficient**

- Ge
- Si

$\lambda$ (nm)

1000 1200 1400 1600 1800

Absorption coefficient (1/cm)

$10^5$ $10^4$ $10^3$ $10^2$ $10^1$

**Schematic - Hybrid**

- Si
- Au
- $E_3\omega$
- $E_\omega$

**SEM image**

**Experimental extinction**

**Simulated extinction**

**E-field distribution**

Experimental extinction (a.u.)

Simulated extinction (a.u.)

Electric energy (a.u.)

Wavelength (nm)

1250 1300 1350 1400

$\lambda = 1325$ nm

$|E| / |E_0|$
Si/Au hybrid – THG


THG – blue emission

THG – blue, green, red emission

Efficiency = 0.007%
We demonstrated that Si nanoantennas can generate the same Raman output signal than Au nanoantennas at $\lambda = 860$ nm, but with 75% less heating.

We obtained a fluorescence enhancement factor of $x3600$ using GaP nanoantennas excited at $\lambda = 633$ nm, originating from high field confinement and strong fluorescence decay acceleration.

By using GaP nanodisks, an enhancement of 3 orders of magnitude in the SHG signal with respect to the bulk was achieved.

We obtained THG efficiency values of up to 0.001% with Ge nanodisks excited at anapole modes, which can also be exploited for FWM phenomena.

We found that the magnitude of the THG emission intensity can be used as a measure of the electric energy within the nanoantenna, and could “observe” the distribution of the E-field by mapping the THG emission.

The hybrid Si/Au nanoantenna was found to produce a THG conversion efficiency of $\sim0.01\%$. The emission maximum could be tuned by changing the size of the antenna.