Theoretical simulation of second harmonic generation from metal-dielectric biresonant nanoantenna

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Second harmonic generation by $BaTiO_3$ particle

$$P_i = \chi_{ij}^{(1)} E_j + \chi_{ijk}^{(2)} E_j E_k + \dots$$

$$E_i = (E_{0i} e^{i(\mathbf{kr}-wt)} + c.c.) \Rightarrow P_i^{(2)} \sim E_0^2 e^{i2(\mathbf{kr}-wt)} + E_0^{*2} e^{-i2(\mathbf{kr}-wt)} + 2E_0 E_0^*$$

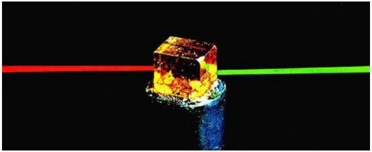
Au: inversion symmetry $\leftrightarrow \chi_{ijk}^{(2)} = 0$
$BaTiO_3$:

$$\begin{pmatrix} P_x^{(2)} \\ P_y^{(2)} \\ P_z^{(2)} \end{pmatrix} = \begin{pmatrix} 0 & 0 & 0 & 0 & \chi_{15} & 0 \\ 0 & 0 & 0 & \chi_{15} & 0 & 0 \\ \chi_{31} & \chi_{31} & \chi_{33} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} E_x^2 \\ E_y^2 \\ E_z^2 \\ 2E_y E_z \\ 2E_x E_z \\ 2E_x E_y \end{pmatrix}$$

(Robet W. Boyd, Nonlinear optics, 2008)

Nonlinear optics applications

Second harmonic generation in macrocrystals is widely used in laser technique.



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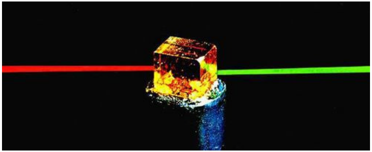
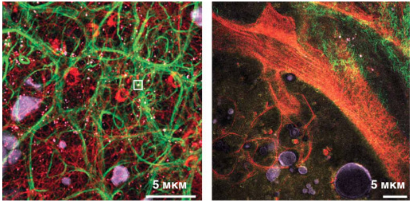
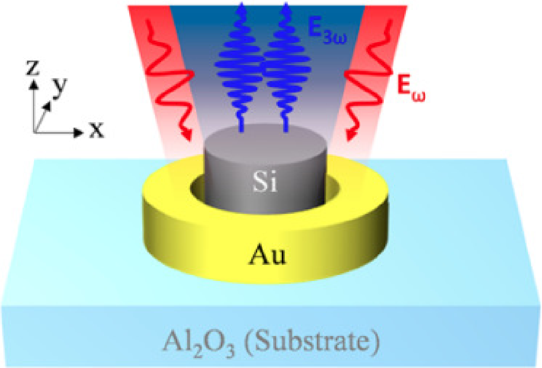


Bioimaging:



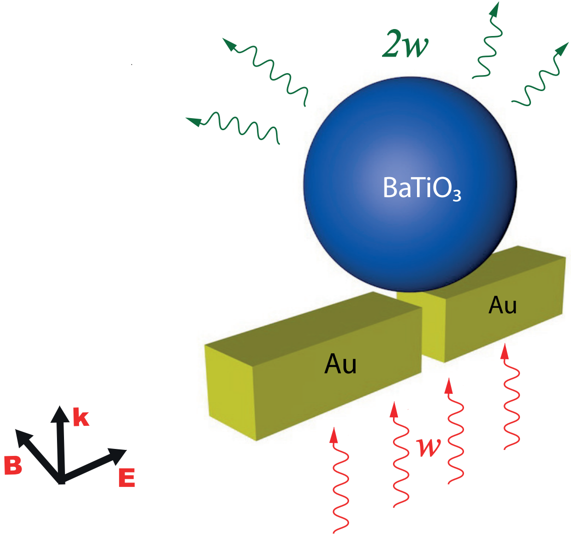
(Delphine Débarre, et. al., Nature, 2005)

Hybrid nanosystems for second harmonic generation



(Toshihiko Shibanuma, et. al., ACS Publications 2017)

Explored system



$\lambda_{inc} = 1200$ нм $\rightarrow \lambda_{scat} = 600$ нм

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Determine nanoantenna configuration for effective second harmonic generation at λ =600 nm wavelength and evaluate generation efficiency.

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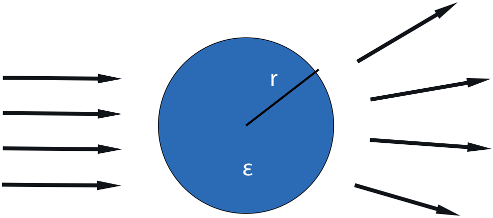
2) Adjust matal nanoparticles for incident field enhancement in infrared region near 1200 nm.
3) Adjust the whole system for incident field enhancement at $\lambda = 1200$ nm and scattered field enhancement on 600 nm. Evaluate qualitatively second harmonic generation by hybrid system and compare it with the case of separate $BaTiO_3$.

$BaTiO_3$ size adjustment for effective second harmonic generation on $\lambda = 600$ nm

Problem № 1



$$\sigma_{scat} = \frac{P_{scat}}{|\mathbf{S}_{inc}|}$$

(S.H. Wemple, et. al., 1968)
$\varepsilon \approx 5$

$BaTiO_3$ size adjustment for effective second harmonic generation on $\lambda = 600$ nm

Electrostatic approximation

$r = 10$ nm

$$\mathbf{p} = r^3 \frac{\varepsilon - 1}{\varepsilon + 2} \mathbf{E}$$

$$\sigma_{scat} \sim (\ddot{p})^2 = -w^2 p^2$$

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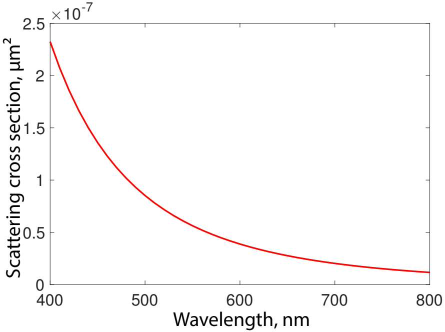
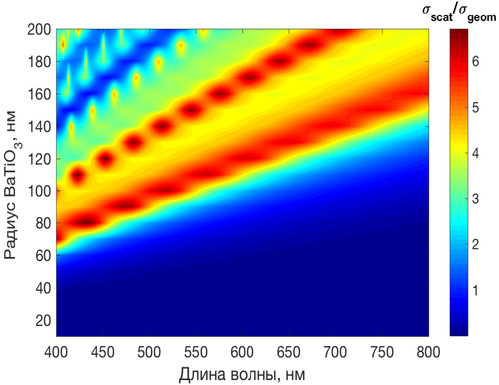


$BaTiO_3$ size adjustment for effective second harmonic generation on $\lambda = 600$ nm

Precise solution (Mie theory): $r \sim \lambda$

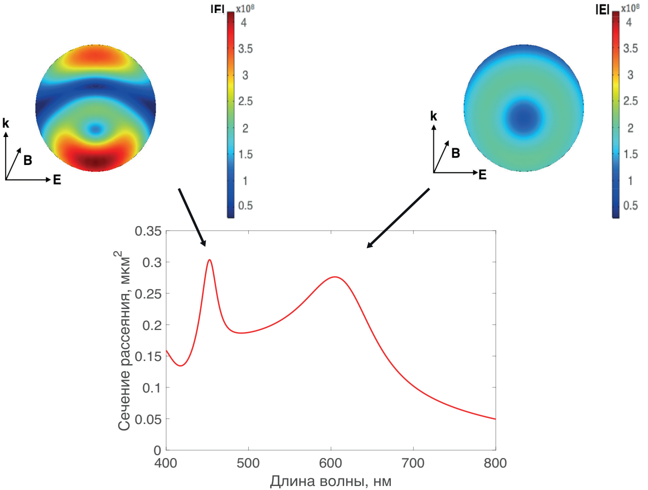
$$\mathbf{E}_s = \sum_{n=1}^{\infty} (a_n M_{e1n}^{(3)} + b_n N_{o1n}^{(3)})$$

$$\sigma_{scat} \sim \sum (|a_n|^2 + |b_n|^2)$$

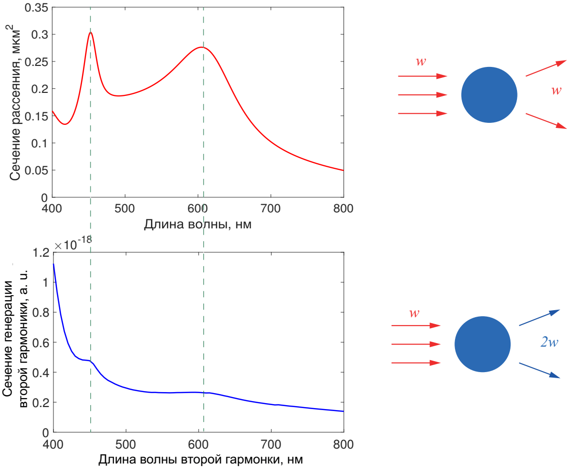


Scattering on $BaTiO_3$ particle with radius r=120 nm

Magnetic quadrupole resonance   Magnetic dipole resonance

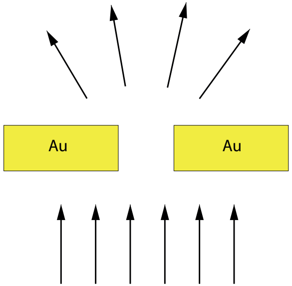


Second harmonic generation by $BaTiO_3$ particle with radius $r = 120$ nm

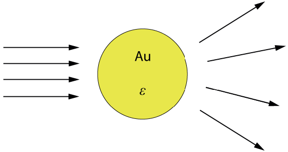


(Robert W. Boyd, "Nonlinear optics 2008)

Problem № 2

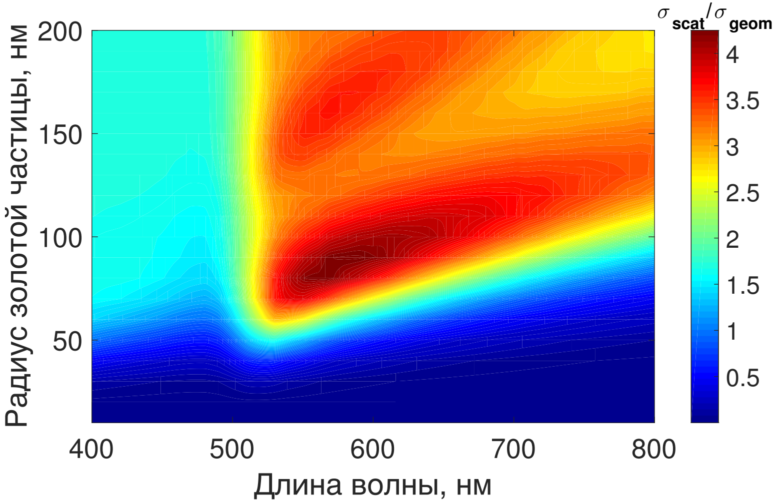


2.1

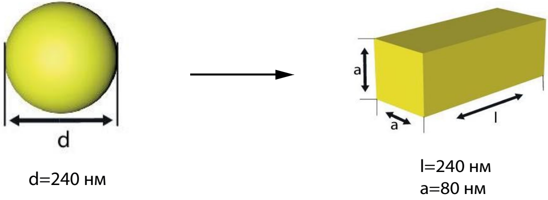


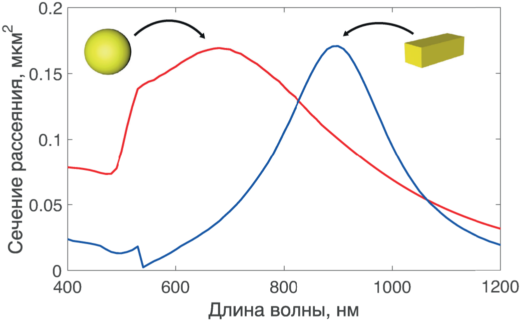
(J. H. Weaver, OSA Publishing, 2015)

Light scattering on gold nanosphere



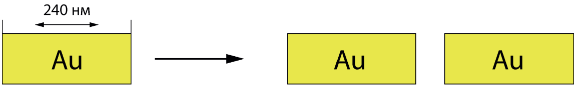
2.2


d
d=240 нм
a
a
l
l=240 нм
a=80 нм

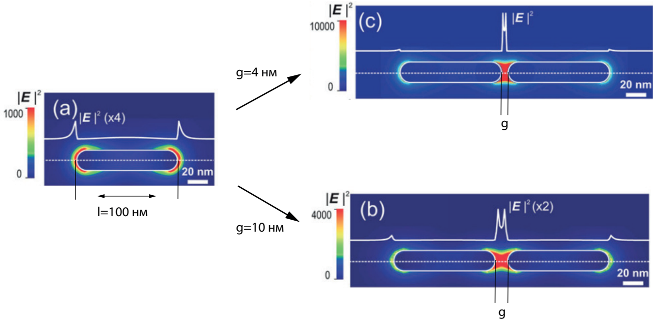

Сечение рассеяния, мкм^2
0.2
0.15
0.1
0.05
0
400
600
800
1000
1200
Длина волны, нм

Light scattering on two gold parallelepipeds

2.3



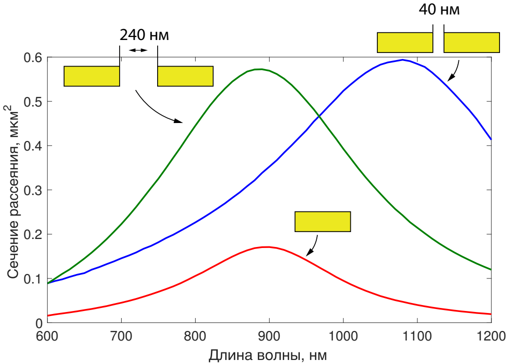
Field enhancement in the gap between parallelepipeds:



(Biagioni P., et. al., NCBI, 2012)

l=240 nm

Scattering cross section dependence on the gap width(g):



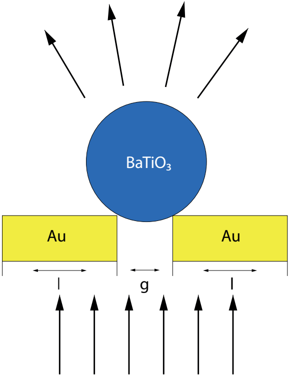
(Biagioni P., et. al., NCBI, 2012)

Scattering cross section dependence on parallelepiped length:

(O. L. Muskens, et al., NCBI, 2007)

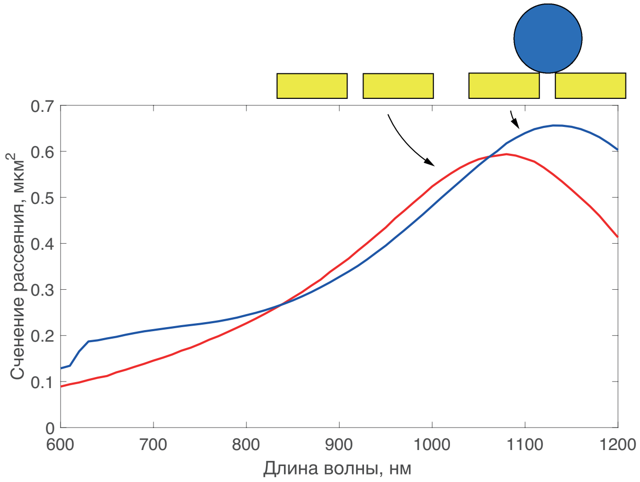
Scattering on the system of $BaTiO_3$ particle and two gold parallelepipeds

Problem № 3

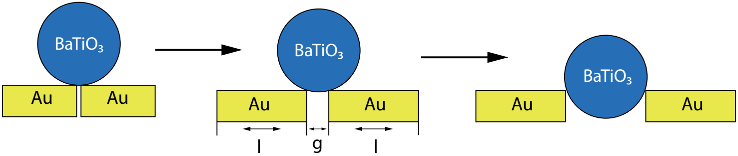


$r(BaTiO_3)=120$ nm
$l=240$ nm; $g=40$ nm

Scattering on the system of $BaTiO_3$ particle and two gold parallelepipeds



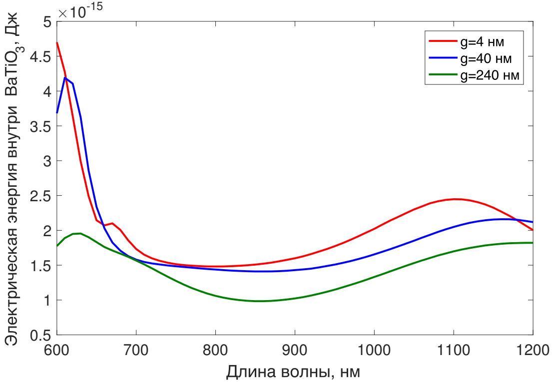
Energy inside $BaTiO_3$ for different configurations



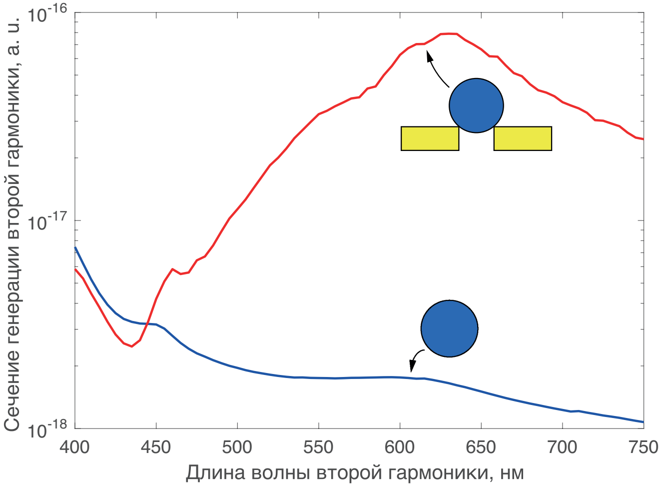
g=4 нм
l=200 нм

g=40 нм
l=240 нм

g=240 нм
l=240 нм



Second harmonic generation by the whole hybrid system



Conclusions

1)$BaTiO_3$ particle optimal size determined for effective second harmonic generation on λ =600 nm wavelength.
2) Gold dimer configuration adjusted for incident field enhancement in infrared region, near λ =1200 nm wavelength.
3) First configuration of the whole system for effective second harmonic generation on λ =600 nm revealed. Demonstrated second harmonic generation enhancement regarding separate $BaTiO_3$ particle.

Thank you for your attention

Second harmonic generation (additional frame)

$$\begin{cases} \mathbf{E}_f \sim \int d\mathbf{r}' \frac{\partial}{\partial t}\mathbf{j}(\mathbf{r}', t - r/c) \\ \mathbf{j} = \mathbf{j}_C + \mathbf{j}_D = \mathbf{j}_C + \dfrac{1}{4\pi}\dfrac{\partial \mathbf{D}}{\partial t} = \mathbf{j}_C + \dfrac{1}{4\pi}\dfrac{\partial \mathbf{E}}{\partial t} + \dfrac{\partial \mathbf{P}}{\partial t} \end{cases}$$