Manipulating Nonlinear Plasmonics by Fundamental Conservation Laws

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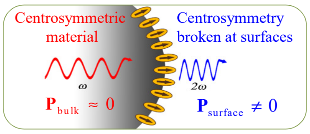
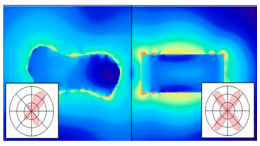
Introduction

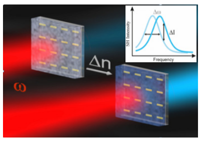
Unique combination of surface-sensitive second harmonic (SH) generation and strong field enhancement in plasmonic structures opening up new possibilities in

- ➢ Ultrasensitive shape characterization
- ➢ Nonlinear plasmonic sensing
- ➢ All optical signal processing
- ➢ High-sensitivity detectors

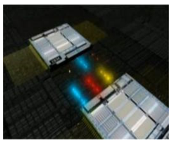




Martin, *et. al.*, Nano Lett. (2013)



Mesch, *et. al.*, Nano Lett. (2016)

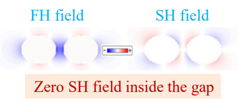


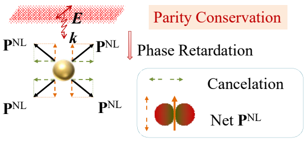
All optical chip, IBM (2011)

Motivation

Weak nonlinear SH process requires strong field enhancement and efficient radiation mechanisms; however, this is full of challenges:

- ➢ Conventional plasmonic enhancement rules breakdown
- ➢ Difficult to control the radiation of SH wave: multipole emission nature
- ➢ Complicated relation between particle shape and resonance
- ➢ Lack of commercial software





- ➢ Parametric nonlinear process governed by fundamental conservation laws
  Charge • Energy • Parity • Angular Momentum

Method

• Coupled-wave Equations for SHG

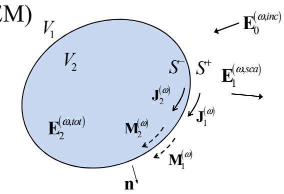
$$\begin{cases} \nabla\times\nabla\times\mathbf{E}^{(\omega)} - \mu_0\varepsilon_0\omega^2\left(1+\chi^{(1)}(\omega)\right)\mathbf{E}^{(\omega)} = i\mu_0\omega\mathbf{J}^{(\omega)} + \underbrace{\varepsilon_0\mu_0\omega^2\chi^{(2)}:\mathbf{E}^{(\Omega)}\mathbf{E}^{(\omega)*}}_{=0} & (1) \\ \nabla\times\nabla\times\mathbf{E}^{(\Omega)} - \mu_0\varepsilon_0\Omega^2\left(1+\chi^{(1)}(\Omega)\right)\mathbf{E}^{(\Omega)} = \varepsilon_0\mu_0\Omega^2\chi^{(2)}:\mathbf{E}^{(\omega)}\mathbf{E}^{(\omega)} & (2) \end{cases}$$

Undepleted-pump Approximation

• Self-consistent Boundary Element Method (BEM)
Initially set $P_S^{(\omega)}$ to be zero, Iteratively solve (1) and (2) to capture mutual coupling.

$$\begin{cases} \boldsymbol{\pi}_0^{(\omega,e)} = -\mathbf{n}\times\mathbf{H}_0^{(\omega,inc)} \\ \boldsymbol{\pi}_0^{(\omega,m)} = \mathbf{n}\times\mathbf{E}_0^{(\omega,inc)} \end{cases} \qquad \begin{cases} \boldsymbol{\pi}_0^{(\Omega,e)} = -i\Omega P_t^S \\ \boldsymbol{\pi}_0^{(\Omega,m)} = \frac{1}{\varepsilon_0}\mathbf{n}\times\nabla_S P_n^S \end{cases}$$

Linear Source Nonlinear Source

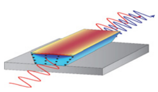


Love's Equivalent Principle

with $\mathbf{P}_S^{(\Omega)} = \varepsilon_0\left[\chi^{(2)}_{\perp\perp\perp}\mathbf{nnn} + \chi^{(2)}_{\perp\|\|}\left(\mathbf{nt}_1\mathbf{t}_1 + \mathbf{nt}_2\mathbf{t}_2\right) + \chi^{(2)}_{\|\perp\|}\left(\mathbf{t}_1\mathbf{nt}_1 + \mathbf{t}_2\mathbf{nt}_2\right)\right]:\mathbf{E}_2^{(\omega)}\mathbf{E}_2^{(\omega)}$

• Self-consistent Maxwell-Hydrodynamic Model

$$\left.\begin{aligned} \nabla\times\mathbf{H} &= \varepsilon_0\frac{\partial\mathbf{E}}{\partial t} + \frac{\partial\mathbf{P}}{\partial t} \\ \nabla\times\mathbf{E} &= -\mu_0\frac{\partial\mathbf{H}}{\partial t}, \end{aligned}\right\}\ \text{Maxwell}$$



$$\left.\begin{aligned} \frac{\partial\mathbf{v}}{\partial t} + \boxed{\mathbf{v}\cdot\nabla\mathbf{v}} &= -\frac{e}{m}\left(\mathbf{E} + \mu_0\boxed{\mathbf{v}\times\mathbf{H}}\right) - \gamma\mathbf{v} - \frac{\nabla p}{n} \\ \frac{\partial n}{\partial t} &= -\nabla\cdot\boxed{(n\mathbf{v})}, \end{aligned}\right\}\ \text{Hydrodynamic}$$

Two systems are coupled through the polarization term

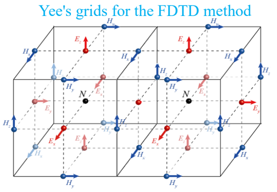
$$\frac{\partial\mathbf{P}}{\partial t} = -en\mathbf{v}.$$

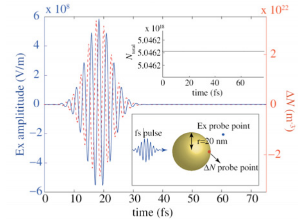
Charge & Energy Conservation

https://doi.org/10.2528/PIER16100401

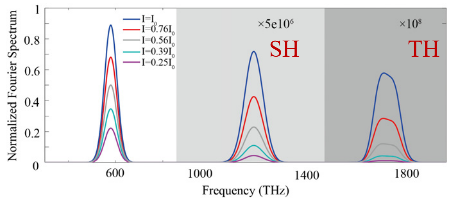
Second & third harmonic generation from a gold nanosphere

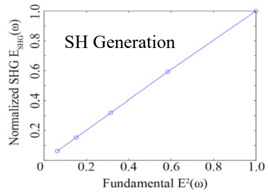
• Electron density fluctuates at each point, the total charge is conserved





• Energy conservation



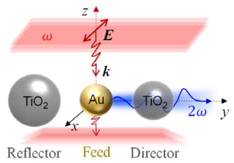


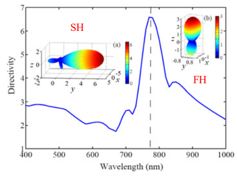
Parity Conservation

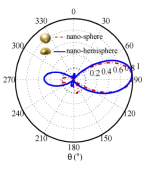
https://doi.org/10.1103/PhysRevA.94.053825
https://doi.org/10.1038/srep18872

Nonlinear nanoantenna design by breaking parity symmetry

• Compact nonlinear Yagi-Uda nanoantenna

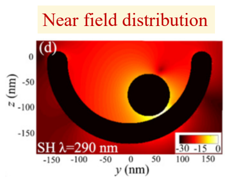


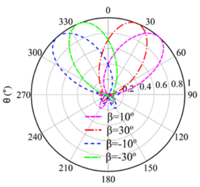


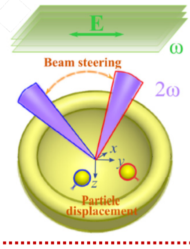


• Particle-in-Cavity Nanoantenna (PIC-NA)

- ➢Cavity: light harvester, concentrator, and reflector
- ➢Strong gap plasmonic enhancement
- ➢Unidirectional radiation by symmetry broken





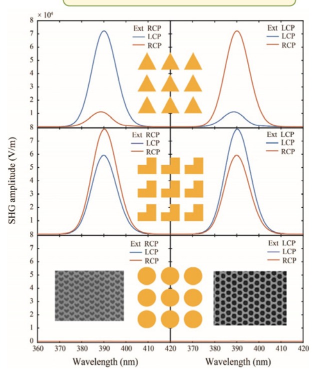


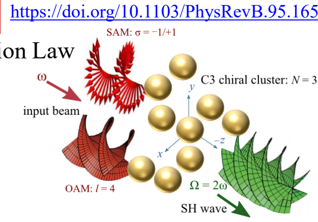
Angular Momentum Conservation

https://doi.org/10.1103/PhysRevB.95.165432

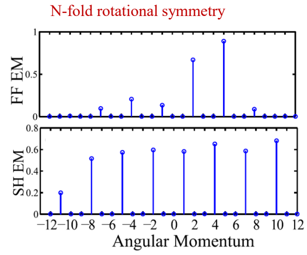
General Angular Momentum Conservation Law

$j_{sca} = s(l+\sigma) + qN$





N-fold rotational symmetry



Supports

AoE/P-04/08