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Second and Third Harmonic Generation at ε-Near-Zero Crossing Point in Arrays of Plasmonic Nanoshells

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Abstract: We theoretically investigate harmonic generation at ε=0 crossing points where losses are compensated through the inclusion of active photonic materials in metallic nanoshells. This singularity-driven process lowers the thresholds for a plethora of nonlinear optical phenomena.
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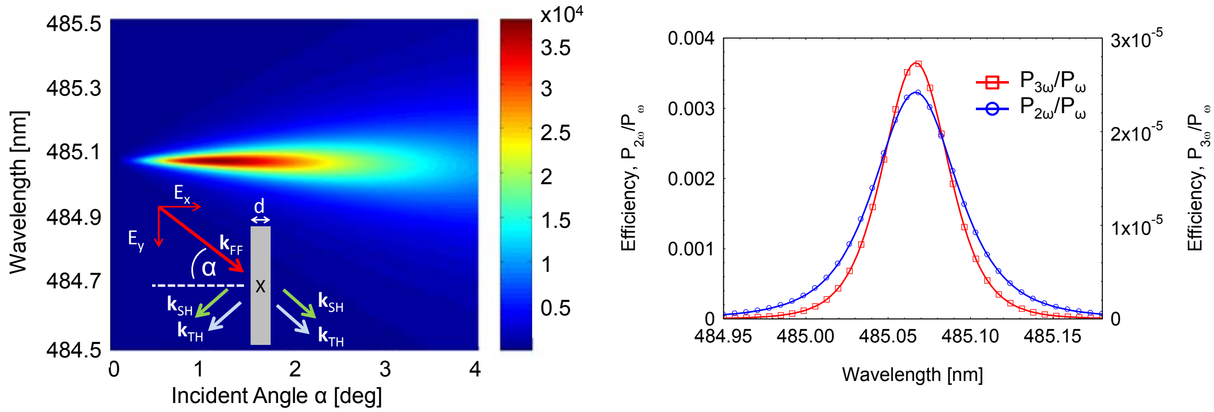
1. Introduction

Renewed attention to surface waves and increased interest in artificial structures [1, 2] have stimulated researchers to study the linear and nonlinear properties of metals and metal-based structures [3, 4]. For example, the strong field enhancement associated with the formation of surface plasmons has led to the exploration of nonlinear optical properties of structures operating in the enhanced transmission regime [4, 5]. Epsilon-near-zero (ε~0) materials have been recently investigated for their peculiar linear [6] and nonlinear optical properties [7, 8]. At the interface between these materials and any other ordinary material (ε≠0), the electric field component orthogonal to the interface becomes singular, due to the requirement of continuity of the displacement field [6-8]. In this paper, we first investigate harmonic generation from a homogeneous layer of a material having a zero-crossing point for the dielectric permittivity in the visible frequency range. We quantify and compare the role of surface and volume nonlinear contributions, which can considerably boost the efficiency of nonlinear processes due to significant electric field enhancement [5, 9]. We then show a practical implementation of such materials, including fluorescent dyes inserted into metallic nanoshells as proposed in reference [10] (though other active photonic materials could be used for this purpose as well). We achieve an effective ε=0 condition in the visible range with a dramatic suppression of losses [10], thus further facilitating the the study of singularity-driven nonlinear optical phenomena.

2. Singularity driven harmonic generation: enhancement via losses compensation

We begin our investigation by considering a generic material *X*, modeled using a single species of Lorentz oscillators that yields the following complex dielectric function $\varepsilon_X(\omega) = 1 - \omega_p^2 / (\omega^2 - \omega_0^2 + i\gamma\omega)$, where the plasma frequency $\omega_p=2\omega_r$, damping $\gamma=10^{-4}\omega_r$, the resonance frequency $\omega_0=0.5\omega_r$, and the reference frequency $\omega_r=2\pi c/1\mu\text{m}$. These parameters produce a strong absorption peak at 2μm and an ε=0 crossing point that displays limited absorption at λ~485nm. The geometry under investigation is depicted in the inset of Fig.1(a): a d=20nm-thick layer of material *X* is illuminated with TM-polarized light (electric field components in the plane of incidence as in Fig.1 (a)). In Fig.1(a) we show that the electric field intensity $|E_x|^2$ inside the material *X* is enhanced approximately 35000 times relative to the incident field intensity ($|E_{inc}|^2=|E_x|^2+|E_y|^2$), and takes on a characteristic tear-drop shape that is symmetric with respect to wavelength, and asymmetric with respect to incident angle α. We then perform the nonlinear calculations in the continuous wave (CW) regime with Comsol Multiphysics [11] and finite difference time domain (FDTD), which allowed us to include surface and volume nonlinear terms. In Fig.1 (b) we report conversion efficiencies for α=1.1°, incident pump intensity 10MW/cm^2, and bulk nonlinearities $\chi^{(2)}$=20pm/V, and $\chi^{(3)}=10^{-21}$(m/V)^2. We observe that harmonic generation correlates directly with the electric field enhancement profile, reaching efficiencies of order 10^{-3} for second harmonic and 10^{-5} for third harmonic.

We note that in this system absorption is not at all negligible, even if Im(ε) may be relatively small: absorption is proportional to the product Im(ε)$|\mathbf{E}|^2$. In our case a reduction of Im(ε) is paired with huge field enhancement, an effect that also leads to more efficient nonlinear processes. For this reason we investigate the possibility to achieve the ε=0 condition and simultaneously abate absorption with the inclusion of active gain materials, e.g. fluorescent dyes, into metallic nanoshells. In particular we model each spherical nanoparticle using the single dipole approximation [10, 12, 13] and the Drude model to describe metal dielectric permittivity. The results reveal that arrays of nanoshells, composed of 10mM of Rhodamine 800 in a silica core with radius r_1=30nm embedded into a gold shell 5nm thick and surrounded by a silica environment, exhibit a ε=0 crossing point around 712nm, with a good overlap of the emission band of the dye and the ε=0 band. At the ε=0 crossing point the loss compensated material exhibits Im(ε)~10^{-4} [10]. We consider a pump signal tuned at ε=0 crossing-point, impinging on a 400nm-thick slab of the above mentioned nanoshells' array at an angle of incidence of α=3.5°. We assume a pump intensity 100MW/cm^2. Under these circumstances, and for bulk $\chi^{(2)}$=0.06pm/V and $\chi^{(3)}$=2.5x10^{-22}m^2/V^2 [15], we estimate an enhancement for the longitudinal component of the electric field intensity inside the metamaterial of ~120 with respect to $|E_{inc}|^2$, and second and third harmonic efficiencies of order 10^{-11}. It is worth stressing that these efficiencies are calculated considering quite small, conservative values of bulk nonlinearities. Moreover in our model we have not considered effective second order nonlinearities arising from symmetry breaking at interfaces, the magnetic Lorentz force [9] or dyes nonlinear contributions. Their inclusion could boost significantly effective second and third order nonlinearities and pave the way for novel low-threshold nonlinear processes.



(a) (b)

Fig.1. (a) Enhancement of the intensity of the electric field component $|Ex|^2$ inside the material *X* with respect to incident electric field intensity $|E_{inc}|^2$ vs wavelength and angle of incidence α (angle formed by the pump wavevector $\mathbf{k}_{FF}$ and the direction orthogonal to the interface). $\mathbf{k}_{SH,TH}$ indicate the wavevectors for second (SHG) and third (THG) harmonic generated signals; (b) Emitted spectra for harmonic fields centered at 242nm and 161nm when α=1.1° vs pump wavelength.

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