ACTIVE BOOSTING OF THE EFFECTIVE OPTICAL NONLINEARITY IN SEMICONDUCTOR NANOMATERIALS

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***Abstract*—** Nanoscale materials usually provide little efficiency as interaction mediums for nonlinear optical processes, such as frequency conversion and parametric amplification, due to their ultra-short length offering little time for considerable wave-mixing. In this paper, it is shown that the effective nonlinear susceptibility of semiconductor nanomaterials can be tuned from ultra-low to gigantic values via controlling the transition carrier density during a given nonlinear optical process. This is achieved by adjusting the intensity of a probe-beam that can be used to control the transition carrier density between the conduction band and the valence band. Since the cross-sectional area and volume of semiconductor nanomaterials are very small, a probe-beam of sufficiently high-power, which has a frequency that is greater than the bandgap frequency, can provide an enormous tuning range for the transition carrier density. Based on numerical simulations, it is shown that within such a huge range, certain values of the transition carrier density can induce a giant nonlinear optical response in nanoscale semiconductors that can be utilized to perform nonlinear operations with the same efficiency as in the macroscale. The attained results are in good agreement with the empirical results for second-harmonic generation efficiency.

Keywords— Harmonic generation, Wave-mixing, Semiconductors, Nanoscale, Optical nonlinearity

I. INTRODUCTION

Nonlinear optical processes are usually quite inefficient in small-scale media due to the ultra-short interaction length of the employed medium [1]. Hence, nonlinear optical operations such as frequency-conversion and parametric amplification are mostly carried out in the macroscale [2]. As nonlinear optical processes are necessary for achieving a high gain-bandwidth product in integrated photonic devices, this hinders the attainment of sufficient optical gain in small-scale optoelectronic devices over a desired bandwidth. Therefore, a drastic increase of the effective nonlinear susceptibility of small-scale optical media, such as optical nanomaterials, is of great interest [3]. In this paper, a numerical investigation on the active tuning of the effective nonlinear susceptibility is carried out for semiconductor nanomaterials via tuning the transition carrier density between the conduction band and the valence band. This can be practically achieved by using a probe-beam of sufficiently high power to excite a huge number of carriers from the valence band to the conduction band owing to the ultra-small volume of semiconductor nanomaterials [4-5]. The probe-beam intensity can then be swept over a large range of values for tuning the transition carrier density and observing the resulting values for the higher order nonlinear susceptibilities, such as the second and the third order susceptibilities, which have practical importance. For the numerical investigation, the electric field wave equation (Eq. 1) [3-4] is concurrently solved with the set of Lorentz dispersion equations for the first and higher order electric polarization densities (Eq. 2-4) [3-4].

$$\nabla^2 E - \mu_0 \varepsilon_\infty \frac{\partial^2 E}{\partial t^2} = \mu_0 \sigma \frac{\partial E}{\partial t} + \mu_0 \frac{d^2 P^{\{3\}}}{\partial t^2} \quad (1)$$

$$\frac{d^2 P^{\{3\}}}{dt^2} + \gamma_r \frac{dP^{\{3\}}}{dt} + {\omega_r}^2 P^{\{3\}} - \frac{{\omega_r}^2 {P^{\{3\}}}^2}{2Ne\zeta} + \frac{{\omega_r}^2 {P^{\{3\}}}^3}{6N^2 e^2 \zeta^2} = \frac{Ne^2 E}{m} \quad (2)$$

$$\frac{d^2 P^{\{2\}}}{dt^2} + \gamma_r \frac{dP^{\{2\}}}{dt} + {\omega_r}^2 P^{\{2\}} - \frac{{\omega_r}^2 {P^{\{2\}}}^2}{2Ne\zeta} = \frac{Ne^2 E}{m} \quad (3)$$

$$\frac{d^2 P^{\{1\}}}{dt^2} + \gamma_r \frac{dP^{\{1\}}}{dt} + {\omega_r}^2 P^{\{1\}} = \frac{Ne^2 E}{m} \quad (4)$$

The effective second and third order nonlinearities ${d^{\{2\}}}_{nl}$ and ${d^{\{3\}}}_{nl}$ can be found by subtracting the lower order polarization densities from the subsequent higher order polarization densities [3-4].

$${P^{\{2\}}}_{nl} = P^{\{2\}} - P^{\{1\}} \quad (5)$$

$${P^{\{2\}}}_{nl} = 2{d^{\{2\}}}_{nl} E^2$$

$${P^{\{3\}}}_{nl} = P^{\{3\}} - P^{\{2\}} \quad (6)$$

$${P^{\{3\}}}_{nl} = 6{d^{\{3\}}}_{nl} E^3$$

The transition carrier density N has to be concurrently solved with Eq. 1-6 through the following rate-equation

$$\frac{dN}{dt} = \Gamma(t) - \frac{N}{\tau_c} = \Gamma(t) - AN - BN^2 - CN^3$$

where the carrier-injection rate Γ is expressed in terms of the probe-beam intensity I (6)

$$\Gamma = \frac{I}{h\nu_{probe}\Delta} = \frac{0.5c\sqrt{\varepsilon_\infty}\varepsilon_0 {E_{probe}}^2}{h\nu_{probe}\Delta}$$

ω_r: $Ang.transition\ frequency$, ε_∞: $Permittivity$

γ_r: $Damping\ rate$, σ: $Conductivity$, c: $Light\ speed$

ζ: $Atom\ diameter$, E: $Electric\ field$, P: $Polarization\ density$

e: $Unit\ charge$, m: $Electron\ mass$ μ_0: $Space\ permeability$

A: $Trap, defect, or\ impurity\ based$

$nonradiative\ recombination\ coefficient$

h: $Planck's\ constant$, τ_c: $Carrier\ lifetime$

B: *Radiative recombination coefficient*, t: *Time*

C: *Auger recombination coefficient*

Δ: *Nanomaterial thickness*, v_{probe}: *Probe frequency*

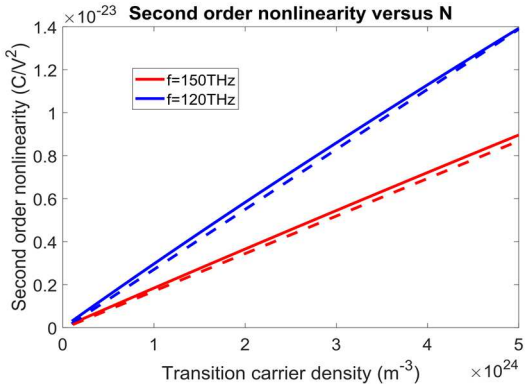

FIG. 1. EMPIRICAL (DASHED-LINE) AND NUMERICAL (SOLID-LINE) VALUES OF THE SECOND ORDER NONLINEARITY (SON) $d^{\{2\}}{}_{nl}$ FOR A 700NM-LONG GAAS, VERSUS N [3,4,6].

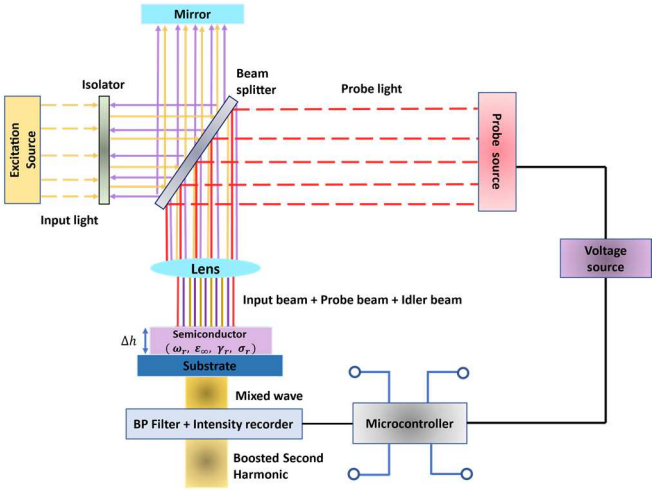

FIG. 2. CONFIGURATION FOR TUNING THE TRANSITION CARRIER DENSITY IN A NANOSCALE SEMICONDUCTOR MEDIUM

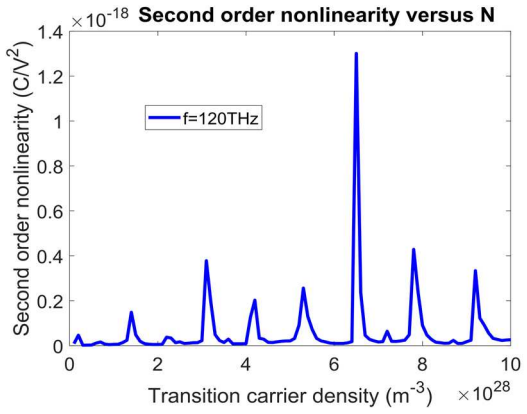

FIG. 3. RESONANCE PEAKS OF THE SON FOR HIGH RANGE OF VALUES OF THE TRANSITION CARRIER DENSITY AT 120THZ.

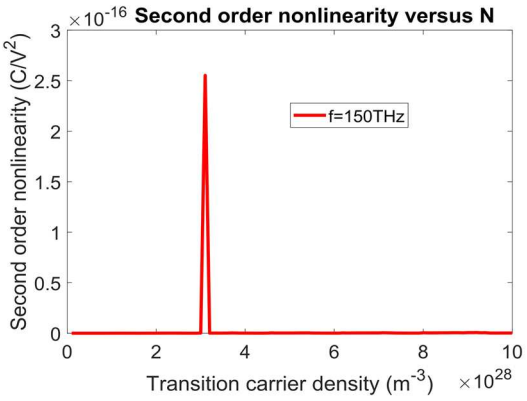

FIG. 4. RESONANCE PEAKS OF THE SON FOR HIGH RANGE OF VALUES OF THE TRANSITION CARRIER DENSITY AT 150THZ.

For the relatively low range of values of N, the resulting SON turned out to be linearly increasing (Fig.1). In the case of a semiconductor nanomaterial (mounted on a substrate) that is excited via a probe-beam of high-power at 120THz using the configuration in Fig.2, the transition carrier density reaches very high values. When the SON is computed for such a high range of values, the resulting variation in SON is observed to be nonlinear and occurs in the form of a series of resonances, where the largest resonance peak occurs at the transition carrier density of $6.6 \times 10^{28} m^{-3}$ with a value of $1.3 \times 10^{-18} C/V^2$. The numerical simulation is repeated for 150THz, in which case a giant resonance peak is detected at $3.2 \times 10^{28} m^{-3}$ with a value of $2.6 \times 10^{-16} C/V^2$. It is discovered that for high range of values of the transition carrier density, the values of the resonance peaks display strong sensitivity to the variation in the beam frequency. Similar resonance peaks occur for higher frequencies with huge ranges of variation, often within $1 \times 10^{-18} C/V^2 < SON < 10^{-15} C/V^2$. Since practical macroscale nonlinear materials usually display an SON within $10^{-24} C/V^2 < SON < 10^{-21} C/V^2$, such a giant boosting of SON in semiconductor nanomaterials enables the efficient performance of nonlinear optical operations such as parametric amplification and harmonic generation in small-scale optoelectronic devices with comparable efficiency.

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