4th URSI AT-RASC, Gran Canaria, 19-24 May 2024

High Harmonic Generation via Cascaded Frequency-Doubling Under Sequential Active-Tuning

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Abstract

This study explores High Harmonic Generation (HHG) via cascaded active frequency-doubling (FD) by employing several stages of optically pumped semiconductor nanofilms. Due to their ultra-small volume, semiconductor nanofilms allow for significant tunability of the transition carrier density. Based on meticulous tuning of the optical pumping rate at each stage, the FD efficiency for an incoming laser beam can be continuously maximized via adaptive optimization of the transition electron density, which greatly enhances the effective medium nonlinearity per-stage. The strategic adjustment of the pump-rate aims to enhance nonlinear light-matter interaction sequentially, paving the way for unprecedented efficiency in HHG.

1. Introduction

High Harmonic Generation (HHG) is a well-known phenomenon in photonics with broad applications in sensing, imaging, and fabrication. The difficulty in achieving HHG arises from the demand for extremely high optical intensity and the fact that only a small fraction of the laser's power contributes to higher harmonics [1,2]. Traditionally, HHG involves focusing powerful ultrashort pulses on tiny spots in gases or on solid surfaces. In our study, we propose an innovative HHG method using a series of identical semiconductor nanofilms for cascaded active frequency-doubling (FD). Due to their ultra-small volume, the semiconductor nanofilms enhance FD efficiency at each step under careful optical pumping that optimizes the transition electron density for high-efficiency FD. Such active tuning can push FD efficiency to nearly 100% per nanofilm, ensuring high intensity for the generated harmonics throughout the cascaded process. This technique achieves an HHG efficiency that surpasses previously reported values [3,4] and can be used for the design of micro and nanoscale HHG devices to be employed in small-scale integrated photonic circuits.

2. Adaptive active tuning of FD efficiency throughout the cascaded process

The process is as shown in Figure 1. The optical pumping rate at each stage is continuously adjusted such that the FD efficiency is adaptively maximized. The number of stages (M) is selected based on the initial laser frequency (ω) and the targeted harmonic ($2^M\omega$). In our computational analysis, the initial frequency of the input wave is set as 300THz, and three stages of 300 nanometers-long Gallium-Arsenide (Ga-As) slabs are modeled for generating an Extreme-UV wave (2400THz).

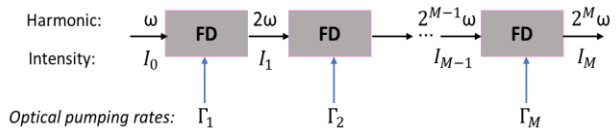

Figure 1. High-harmonic generation through cascaded FD based on optical active-tuning.

3. Formulation

The implementation is carried out via a simultaneous solution of the electric field wave equation and the associated nonlinear Lorentz dispersion equation, to model the wave propagation and the dispersion characteristics of the Ga-As nanofilms. The electron dynamics within each Ga-As nanofilm is modeled by the semiconductor injection-rate equation. The input wave electric field is excited at a given point in one-dimensional space. Assuming that M identical Ga-As nanofilms are used in the cascaded system (i=1,2,...,M), one needs to solve the following set of equations within each nanofilm [4]

Electric field wave equation:

$$\nabla^2 E_i(f_i) - \mu_0 \varepsilon_\infty \frac{\partial^2 E_i(f_i)}{\partial t^2} = \mu_0 \sigma \frac{\partial E_i(f_i)}{\partial t} + \mu_0 \frac{d^2 P_i}{dt^2} \quad (1)$$

Nonlinear Lorentz dispersion equation:

$$\frac{d^2P_i}{dt^2}+\gamma\frac{dP_i}{dt}+{\omega_0}^2P_i-\frac{\left(\frac{1}{2}\right){\omega_0}^2{P_i}^2}{(N_0-N_i)ed}+\frac{\left(\frac{1}{6}\right){\omega_0}^2{P_i}^3}{(N_0-N_i)^2e^2d^2}=\frac{(N_0-N_i)e^2E_i(f_i)}{m_e} \quad (2)$$

Injection − rate equation:

$$\frac{dN_i}{dt}=\Gamma_iN_i-\frac{N_i}{\tau_{e,i}},\ \tau_{e,i}=\frac{1}{A+BN_i+C{N_i}^2},\ \text{where } \Gamma_i=\frac{I_i}{h\nu\Delta w} \quad (3)$$

FD efficiency:

$$\eta_{FD,i}=\frac{\text{Intensity of the second harmonic at } x=x_{out,i}}{\text{Intensity of the first harmonic at } x=x_{in,i}}=\frac{\left|\int_0^{\Delta T}\{E\,(x=x_{out,i},t)\,e^{-i(4\pi f_i)t}\}dt\right|^2}{\left|\int_0^{\Delta T}\{E\,(x=x_{in,i},t)\,e^{-i(2\pi f_i)t}\}dt\right|^2} \quad (4)$$

τ_e: *Carrier lifetime*, I: *Pump intensity*, Γ: *Optical pumping rate*, η_{FD}: *FD efficiency*

E: *Electric field*, μ_0: *Free space permeability*, ε_∞: *Background permittivity*, t: *time*, h: *Planck's constant*, η_{FD}: *FD efficiency*

σ: *Conductivity*, P: *Polarization density*, γ: *Polarization decay rate*, ΔT: *Pulse duration*

ω_0: *Angular resonance frequency*

N_0: *Bound electron density*

ω: *Angular frequency*, N: *Free electron density*, e: *Elementary charge*, d: *Atom diameter*

m_e: *Electron mass*, v: *Pump frequency*

Δw: *Longitudinal material thickness*

f_i: *Input frequency for the ith segment*

A: *Trap, impurity, or defect based recombination coefficient*, B: *Radiative recombination coefficient*, C: *Auger recombination coefficient*

For each stage (within each nanofilm), Equations 1-4 are iteratively solved via Equation 5 such that the optical pumping rate yields out the maximum FD efficiency,

$$\Gamma_i^{(k)}=\Gamma_i^{(k-1)}-\alpha_k\left(\boldsymbol{H}^{(k-1)}\right)^{-1}\left(\boldsymbol{\nabla}\eta_{FD,i}\left(\Gamma_i^{(k-1)}\right)\right),\ k=1,2,3,\ldots \quad (5)$$

$\eta_{FD,i}\left(\Gamma_i^{(k-1)}\right)$: *Objective function*, $\Gamma_i^{(k)}$: *Optimization parameter*, $\boldsymbol{H}^{(k-1)}$: *Hessian matrix*, α_k: *Step size*

The Hessian matrix is evaluated according to the BFGS algorithm [4] and the step size is determined based on *Wolfe's conditions* at each iteration for a given stage. By maximizing FD efficiency at each stage, the algorithm retains the amplitude level of the electric field (incoming laser beam) until the intended harmonic is generated at the desired intensity. As displayed in Figure 3, the resulting magnitude of the final harmonic (Extreme-UV) at 2400THz can be nearly equal to the initial harmonic at 300THz (Near-IR) as a consequence of continuous active tuning, which is very difficult to achieve using the conventional method [5]. Our numerical FD results agree with the experimental ones [6]. The presented algorithm can be used to achieve efficient HHG in micro and nanoscale optical devices for employment in functional small-scale integrated circuits.

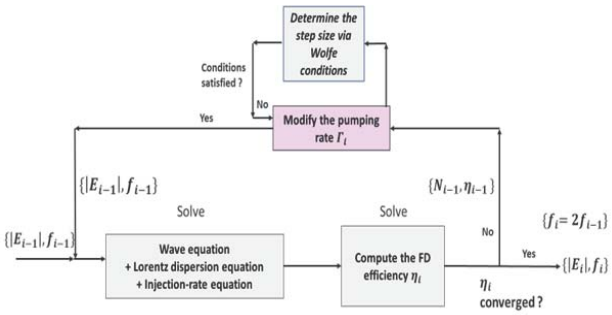

Figure 2. Algorithm for High-Harmonic Generation through cascaded active Frequency-Doubling.

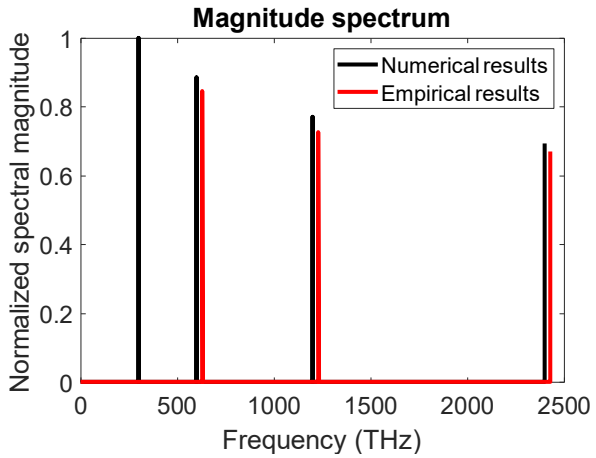

Figure 3. The resulting spectrum via High-harmonic generation through three-stage active frequency-doubling.

References

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