Nonlinearity Analysis of 2D Materials by Using GS-FDTD Method

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***Abstract*—We present a new numerical method to study the linear and nonlinear response of dispersive and nonlinear 2D materials by incorporating a nonlinear generalized source (GS) into the finite-difference time-domain (FDTD) method. This new method is particularly powerful when applied to the analysis of 2D materials, as most such materials cannot be studied directly by traditional FDTD method due to their vanishingly small thickness. A typical graphene grating has been studied to verify the accuracy of the proposed GS-FDTD method, and its predictions agree well with other well-known frequency-domain numerical methods.**

***Index Terms*—nonlinearity, nonlinear generalized source (GS), SHG, THG, FDTD, 2D materials.**

I. Introduction

Owing to its unique and novel linear optical properties, two-dimensional (2D) materials have been widely used in practical applications [1]-[3], including nanolasers, sensors, solar cells, and optical modulators. In addition to the outstanding linear property, the nonlinear property of 2D materials, particularly the second-harmonic generation (SHG) and third-harmonic generation (THG), play an equally important role in optical regime [4]-[6]. For example, some typical 2D materials (such as graphene and transition-metal dichalcogenides) are already used to design high-performance nanoscale frequency mixers and other active optoelectronic devices. However, these promising applications present serious challenges for traditional FDTD method [7]. Different from the numerical analysis of strong nonlinearity caused by multipactor in high-power microwave devices [8][9], if we want to study the extremely week SHG and THG by using FDTD method, we have to solve complex, computationally intensive time-domain convolution integrals [10][11]. Such integrals generally require large computational time and memory, and these computational resources increase rapidly upon marching the algorithm in time. To avoid these problems, currently the FDTD method is mostly used for instantaneous, dispersionless nonlinear effects, such as the Kerr effect. However, in the study of optical nonlinearities of 2D materials, one is primarily interested on the second- and third-harmonic generation, as these nonlinear optical effects are used in many active photonic nanodevices. An additional challenge comes from the fact that second- and third-order nonlinear susceptibilities are usually dispersive and anisotropic. In order to overcome these challenges and be able to study the SHG and THG in 2D materials, we developed a new method described here, namely a GS-FDTD that integrates the GS and FDTD methods.

II. Algorithm

From the definitions [12] of second- and third-harmonic generation, it is known that the intensity of SHG and THG is determined only by the local field at the fundamental frequency (FF). Moreover, compared to other nonlinear processes, the SHG or THG are by far the most studied as many technological applications rely on these nonlinear optical interactions. Importantly in this context, we can study SHG or THG with only two different linear FDTD simulations. The first linear simulation consists of exciting the system by a regular, linear source, whereas in the second simulation the system is excited by a nonlinear generalized source (GS). The basic steps of the implementation of the GS-FDTD method are summarized in what follows.

Step 1: Linear simulation at fundamental frequency

In the first linear FDTD simulation, we assume there are only linear materials in the computational region, and stimulate this linear system with a linear source at the FF. As a result, we can calculate the time-domain (TD) near-field distribution at FF by using one single FDTD simulation.

Step 2: Nonlinear generalized source evaluation

Different from the first linear simulation, the second linear simulation at high-order frequencies is excited with a nonlinear generalized source. Such nonlinear generalized source [4][5] for the THG of graphene is evaluated as

$$J_i^{(3)}(3\omega,\omega)=\sigma_s^{(3)}(3\omega,\omega)\sum_{jkl}\Delta_{ijkl}^{(3)}E_j(\omega)E_k(\omega)E_l(\omega) \quad (1)$$

where, the subscripts (i,j,k,l) represent the components (x,y,z) individually. $\Delta_{ijkl}^{(3)}=\left(\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}\right)/\left(3h_{eff}\right)$ and δ_{ij} is the Kronecker delta. As the nonlinear surface current lies onto the plane of 2D materials, the value of the normal component of current density in (1) is zero.

In order to compute the frequency-domain (FD) near-field in (1), the TD near-field distribution at FF obtained at Step 1 is first Fourier transformed to the frequency domain. Subsequently, we use (1) to evaluate the FD nonlinear current density. To incorporate these FD nonlinear current sources into the FDTD simulation, we inverse Fourier transform them to obtain the corresponding TD nonlinear current sources. Here, we note that the number of frequency sampling points should satisfy Nyquist-Shannon sampling theorem strictly, so that the final TD nonlinear current source can be calculated accurately via the inverse Fourier transformation.

Step 3: Linear simulation at high-order frequency

In the second linear FDTD simulation, we can assume that there are only linear materials. However, unlike the procedure in Step 1, the second linear FDTD simulation is excited with the time-dependent nonlinear current source obtained at Step 2 rather than an external excitation. In this way, we can accurately calculate the intensity of SHG and THG, and in fact any other instantaneous nonlinear optical process. Compared to the nonlinear frequency-domain method [13][14], the FD electric field at different frequencies in (1) can be obtained from a single FDTD simulation via Fourier transformation, rather than repeating the FD simulation for each frequency. Thus, GS-FDTD is usually computationally less demanding and more efficient than nonlinear FD methods.

III. Results and Discussion

The proposed GS-FDTD method is a general numerical method to study the SHG and THG in 2D materials. In order to illustrate its versatility and efficiency, we investigate here the double resonance phenomenon in graphene nanostructures [15]. As shown in Fig.1, our photonic structure consists of a periodic graphene grating with the ribbons oriented along the *X* axis. The period in this example is Λ=100 nm, and the width of the ribbons, *W*=86 nm. The graphene grating is placed in the *XY* plane, and its chemical potential is 0.6 eV. The relaxation time is 0.25 ps, and the temperature is T=300 K.

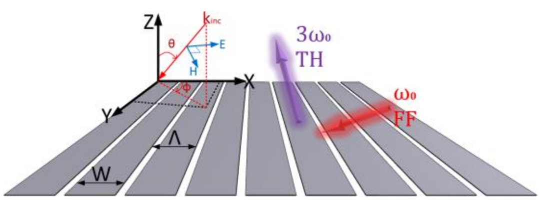

Fig. 1: Schematics of a graphene grating with period Λ and width *W* .

In the first linear FDTD simulation, the graphene grating is illuminated by a plane wave with a Gaussian pulse envelope. This pulse covers the whole fundamental-frequency range [30-150 THz]. The incident angles in Fig.1 are $\theta = 0$ and $\phi = 0$. Based on its FF near-field distribution, we use the proposed GS-FDTD method to calculate the nonlinear response of this graphene grating, and the corresponding nonlinear results are given in Fig. 2. Similar to the linear case, there are a series of strong peaks in Fig. 2a, the near-field profiles for the first three peaks being depicted by Figs. 2b through 2d. In addition, we have compared our results with the well-known GS-RCWA method [14][15] in Fig. 2a, which shows that they agree very well. This verifies the accuracy of the proposed GS-FDTD.

As we known, GS-RCWA is a specialized method, which is highly efficient for several particular applications. But its major challenge is that it is limited to model periodic structures, and just applicable primarily to diffraction problems. By contrast, the proposed GS-FDTD method is a general-purpose numerical method. In particular, it can be utilized to model not only periodic structures, but also devices of finite extent. More importantly, in addition to the diffraction problem just discussed, GS-FDTD method can be used to study much more complicated nonlinear problem, such as light propagation in a nonlinear medium in non-paraxial approximation, design of high-Q nonlinear photonic crystal cavities, and radiation from particle clusters. Therefore, GS-FDTD method is more general and suitable for study of nonlinear optical phenomena.

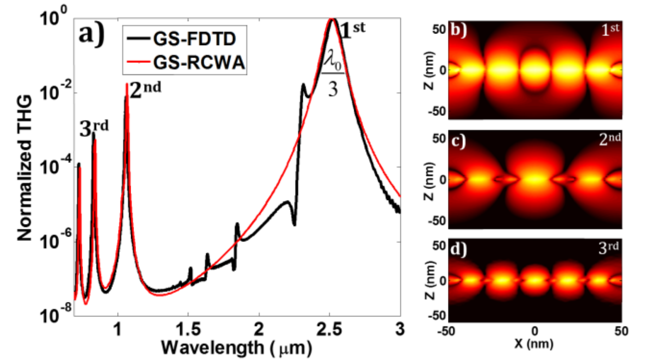

Fig. 2: Nonlinear results at TH: (a) Comparison of THG calculated by different methods and (b)-(d) Field profile of |Ex| for the first three resonance modes in Fig. 2a.

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