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The effect of higher order harmonics on second order nonlinear phenomena

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ABSTRACT

A new method which is a combination of the harmonic balance and finite difference techniques (HBFDF) is proposed for complete time-harmonic solution of the nonlinear wave equation. All interactions between different harmonics up to an arbitrary order can be incorporated. The effect of higher order harmonics on two important nonlinear optical phenomena, namely, the second harmonic generation (SHG) and frequency mixing is investigated by this method and the results are compared with well-known analytical solutions. The method is quite general and can be used to study wave propagation in all nonlinear media.

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1. Introduction

Since the invention of laser, propagation of high intensity optical waves which is substantially affected by the nonlinear properties of the medium has been a topic of high interest. Non-linearities of the medium lead to optical phenomena such as second harmonic generation, frequency mixing, self-refraction, self-phase modulation and soliton which have all found interesting applications in optoelectronics and optical communications.

In order to study these phenomena, the wave equation must be solved in nonlinear (NL) media. Approximate and simplified closed-form solutions of the NL wave equation for some of these phenomena already exist. These solutions are usually obtained by neglecting higher order harmonics and employing other simplifying assumptions such as slowly varying envelope approximation (SVEA) [1,2] which are only applicable when the nonlinear effects are very weak [3].

Nonlinear Schrödinger equation (NLSE) has been widely used for Soliton propagation and SHG [4–6]. Existence of a slowly varying envelope is the fundamental assumption behind the derivation of NLSE. On the other hand, purely numerical techniques such as finite difference time domain (FDTD) and beam

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propagation method (BPM) have also been used to study a number of nonlinear problems such as SHG, self-focusing, and Soliton propagation [7–19]. In conventional BPM such as FFT-BPM [15] or FD-BPM [16] the linear and nonlinear parts of the paraxial scalar wave equation are treated separately. In Bidirectional BPM an iterative procedure is employed which is started by solving an independent linear problem to calculate the input/output components of the electric field. Then, energetic exchanges between the two harmonics, which are due to the NL characteristic of the medium, are computed by using these components. Finally, the input/output field components are calculated by considering the energetic exchanges. This iteration stops when the difference between new and old components becomes less than a predefined tolerance [20,21]. Increasing the number of harmonics in this method quickly increases the complexity in calculation of the total energetic exchanges. FDTD can yield more accurate results than those of conventional BPM because it does not use the paraxial approximation. On the other hand, in order to minimize the effects of numerical dispersion while maintaining stability, FDTD requires fine temporal and spatial discretizations which leads to high computational cost of this method [7,8].

In this paper, a new time-harmonic solution for the nonlinear wave equation is presented in which the effects of higher order harmonics up to an arbitrary order are included. The proposed method is inspired by the Harmonic Balance Technique (HBT) which is a well-known method for the analysis of lumped and distributed nonlinear circuits [22]. In this method, all interactions among different harmonics are taken into account while the number of harmonics involved is limited by the user. HBT has also

been applied to the analysis of nonlinear transmission lines with periodic excitation and formation of shockwaves and solitons in such structures have been reported [23]. The common practice is that the nonlinear transmission line is divided into small segments and each segment is replaced by a lumped element equivalent circuit with nonlinear capacitors. The resulting lumped element circuit is then solved by HBT. The number of nonlinear elements depends on the electrical length of the transmission line, consequently, analyzing a long nonlinear transmission line can become very time consuming by this approach. To overcome this problem, our proposed technique uses finite difference method to solve the nonlinear differential equation in frequency domain. First, the solution is expanded in terms of multiple temporal harmonics with spatially varying coefficients. After balancing the harmonics, a system of nonlinear differential equations for the coefficients is obtained which is solved by the finite difference method. Finally, the Manley–Rowe relations are used to check the balance of power in the medium [24]. The proposed method is called HB-FD technique. It will be used to simulate SHG and frequency mixing in one-dimensional lossless nonlinear media by considering the effects of higher order harmonics. It will be shown that the presence of a higher order harmonic will strongly influence both nonlinear phenomena. It should be stressed that simplifying assumptions such as paraxial approximation and SVEA are not used in the proposed formulation, therefore, not only strong nonlinearity can be considered but the true phase mismatch is also calculated based on the actual dispersion characteristics of the medium. Here the linear and NL parts are not separated and any number of harmonics or combination of waves at different frequencies can be easily incorporated into the solution procedure. These characteristics result in lower complexity, faster solution, and more versatility compared to Bidirectional BPM [20]. Unlike FDTD which can be used to simulate the propagation of narrow pulses of light, the proposed method in its current form can only handle a superposition of finite number of monochromatic waves with different frequencies. Compared to similar methods that employ HBT for modeling wave propagation in nonlinear transmission lines [25], HB-FD technique is faster and can be easily adapted for lossy, dispersive, or inhomogeneous media.

2. Formulation

2.1. Nonlinear wave equation

The wave equation in a NL, homogeneous, and anisotropic medium is derived simply from Maxwell's equations [26]:

$$\nabla^2 \mathbf{E} - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}}{\partial t^2} \quad (1)$$

in which $c = 1/\sqrt{\epsilon_0 \mu_0}$ is the speed of light in vacuum. The polarization vector \mathbf{P} can be separated into linear and NL parts:

$$\mathbf{P} = \epsilon_0 \chi^{(1)} \mathbf{E} + \epsilon_0 (\overline{\chi}^{(2)} : \mathbf{E}^2 + \overline{\chi}^{(3)} : \mathbf{E}^3 + \dots) = \mathbf{P}_L + \mathbf{P}_{NL} \quad (2)$$

where $\chi^{(1)}$ is the linear first order susceptibility, and $\overline{\chi}^{(2)}$, $\overline{\chi}^{(3)}$, ... are NL higher order susceptibilities [1,4]. When the NL response of the medium is not instantaneous, the successive terms in the above equation should be replaced by convolutions in time domain [26].

2.2. Solution of NL wave equation by HB technique

In this section harmonic balance technique is used to obtain a steady state time-harmonic solution for the NL wave equation. The electric field and polarization are expanded in terms of multiple

temporal harmonics with spatially varying coefficients:

$$\mathbf{E} = \sum_{n=1}^{\infty} \mathbf{E}_n(\mathbf{r}) e^{j\omega_n t} \quad (3)$$

$$\mathbf{P} = \sum_{n=1}^{\infty} \mathbf{P}_n(\mathbf{r}) e^{j\omega_n t} \quad (4)$$

where ω_n stands for various frequencies which include fundamental frequencies, their integer harmonics, and linear combinations of them. \mathbf{E}_n and \mathbf{P}_n are complex vector coefficients of the electric field and polarization at frequency ω_n which, in general, are functions of spatial coordinates. Substituting (3) and (4) into (1) yields

$$\sum_{n=1}^{\infty} \left[\nabla^2 \mathbf{E}_n(\mathbf{r}) + \frac{\omega_n^2}{c^2} \mathbf{E}_n(\mathbf{r}) + \mu_0 \omega_n^2 \mathbf{P}_n(\mathbf{r}) \right] e^{j\omega_n t} = 0 \quad (5)$$

After substituting frequency domain descriptions of linear and NL polarizations into (5) and only considering NL susceptibilities of the second and third order (higher order NL susceptibilities are usually insignificant and neglected) we obtain [4]

$$\begin{aligned} & \sum_{n=1}^{\infty} \left[\nabla^2 \mathbf{E}_n + \frac{\omega_n^2}{c^2} (1 + \chi^{(1)}(\omega_n)) \mathbf{E}_n \right] e^{j\omega_n t} \\ & + \sum_{n=1}^{\infty} \frac{\omega_n^2}{c^2} \sum_k \sum_l \overline{\chi}^{(2)}(\omega_n; \omega_k, \omega_l) : \mathbf{E}_k \mathbf{E}_l e^{j\omega_n t} \\ & + \sum_{n=1}^{\infty} \frac{\omega_n^2}{c^2} \sum_p \sum_q \sum_r \overline{\chi}^{(3)}(\omega_n; \omega_p, \omega_q, \omega_r) : \mathbf{E}_p \mathbf{E}_q \mathbf{E}_r e^{j\omega_n t} = 0 \end{aligned} \quad (6)$$

In order for (6) to hold at all times, coefficients of $\exp(j\omega_n t)$ must be zero. Hence, a NL system of equations is derived which is the time-harmonic equivalent of the NL wave equation:

$$\begin{aligned} \nabla^2 \mathbf{E}_n + \frac{\omega_n^2}{c^2} (1 + \chi^{(1)}(\omega_n)) \mathbf{E}_n + \frac{\omega_n^2}{c^2} \sum_k \sum_l \overline{\chi}^{(2)}(\omega_n; \omega_k, \omega_l) : \mathbf{E}_k \mathbf{E}_l \\ + \frac{\omega_n^2}{c^2} \sum_p \sum_q \sum_r \overline{\chi}^{(3)}(\omega_n; \omega_p, \omega_q, \omega_r) : \mathbf{E}_p \mathbf{E}_q \mathbf{E}_r = 0 \\ n = 1, 2, 3, \dots \end{aligned} \quad (7)$$

To demonstrate the basic steps of the proposed method, in this paper, we only consider plane-wave propagation in an unbounded isotropic NL medium. It is assumed that the plane-wave propagates along the z -axis and the electric field has a linear polarization along the x -axis. The medium is homogeneous in transverse plane but its constitutive parameters may vary along the direction of propagation. Therefore, (7) is reduced to a scalar equation:

$$\begin{aligned} \frac{d^2 E_n}{dz^2} + \frac{\omega_n^2}{c^2} (1 + \chi^{(1)}(\omega_n)) E_n + \frac{\omega_n^2}{c^2} \sum_k \sum_l \chi^{(2)}(\omega_n; \omega_k, \omega_l) E_k E_l \\ + \frac{\omega_n^2}{c^2} \sum_p \sum_q \sum_r \chi^{(3)}(\omega_n; \omega_p, \omega_q, \omega_r) E_p E_q E_r = 0 \\ n = 1, 2, 3, \dots \end{aligned} \quad (8)$$

in which $E_n(z)$ is the electric field component of the n th harmonic and $\chi^{(i)}$ may be function of z if the medium is inhomogeneous. The system of second order NL differential equations in (8) must be solved numerically.

2.3. Finite difference method

The finite difference method can be used to solve the system of NL differential equations in (7) or (8) in space domain [27]. To solve (8), the z -axis between $z=0$ and $z=L$ (L is arbitrary) is

divided into M equal segments of length $\Delta z = L/M$. At each point $z_i = i\Delta z$, $i = 1, 2, 3, \dots, M-1$ the central difference approximation is used to discretize the second order derivatives in (8). Consequently, a set of nonlinear algebraic equations is obtained which must be solved after the initial and/or boundary conditions are imposed. Although this solution produces the forward and backward waves in the medium, the number of harmonics used in the solution and also the number of discrete points along the medium might lead to a very large system of nonlinear equations. In order to overcome this problem, the medium is assumed to be semi-infinite and any reflection from $z=L$ is ignored as in [28]. This assumption enables us to use the following equation to update the electric field at each point using its values at previous points:

$$E_n^{i+1} = 2E_n^i - E_n^{i-1} - (\Delta z)^2 \frac{\omega_n^2}{c^2} \left[(1 + \chi_i^{(1)}(\omega_n)) E_n^i + \sum_k \sum_l \chi_i^{(2)}(\omega_n; \omega_k, \omega_l) E_k^i E_l^i + \sum_p \sum_q \sum_r \chi_i^{(3)}(\omega_n; \omega_p, \omega_q, \omega_r) E_p^i E_q^i E_r^i \right] \quad (9)$$

$n = 1, 2, 3, \dots \quad i = 1, 2, 3, \dots, M-1$

where the superscript (subscript) i means that the quantity is computed at $z = i\Delta z$. E_n^0 and E_n^1 are initial values of the electric field of n th harmonic at $z = 0$, Δz when the electromagnetic wave enters the NL medium. Note that the recursive finite difference method does not require any boundary condition at $z = M\Delta z = L$, in other words, there is no reflection at this point because the medium is semi-infinite. In the following sections after computing the electric field of each harmonic, in order to plot the envelope variations of the field we will separate the envelope and phase variations of E_n defined by

$$E_n(z) = A_n(z)e^{-jk_n z} + B_n(z)e^{jk_n z} \quad (10)$$

in which $A_n(z)$ and $B_n(z)$ represent the forward and backward electric field envelope variation, respectively. k_n is the phase constant at frequency ω_n , in a non-dispersive medium k_n equals $\omega_n \sqrt{\mu\epsilon}$. Here, $B_n(z)$ is assumed to be zero as there is no reflection in a homogeneous semi-infinite medium.

3. Numerical results

3.1. Second harmonic generation

Wave propagation in NL media with asymmetric constructive crystals, in which $\chi^{(2)}$ is nonzero and dominant, leads to the generation of second harmonic (SH) frequency component [1]. This phenomenon is known as the second harmonic generation (SHG). In such media, the NL polarization in (2) is well approximated by $\epsilon_0 \chi^{(2)} E^2$. Consequently, Eq. (9) is further simplified when only the fundamental frequency (FF) and its second harmonic, $2\omega_0$, are considered to be propagating through the medium:

$$E_{\omega_0}^{i+1} = 2E_{\omega_0}^i - E_{\omega_0}^{i-1} - (2\pi\Delta z')^2 \left[(1 + \chi^{(1)}(\omega_0)) E_{\omega_0}^i + 2\chi^{(2)}(\omega_0; 2\omega_0, -\omega_0) E_{2\omega_0}^i E_{\omega_0}^i \right] \quad (11)$$

$$E_{2\omega_0}^{i+1} = 2E_{2\omega_0}^i - E_{2\omega_0}^{i-1} - (2\pi\Delta z')^2 \left[4(1 + \chi^{(1)}(2\omega_0)) E_{2\omega_0}^i + 4\chi^{(2)}(2\omega_0; \omega_0, \omega_0) E_{\omega_0}^i E_{\omega_0}^i \right] \quad (12)$$

where $\Delta z' = \Delta z/\lambda_0$, $\omega_0/c = 2\pi/\lambda_0$, and E_n^i is the complex conjugate of

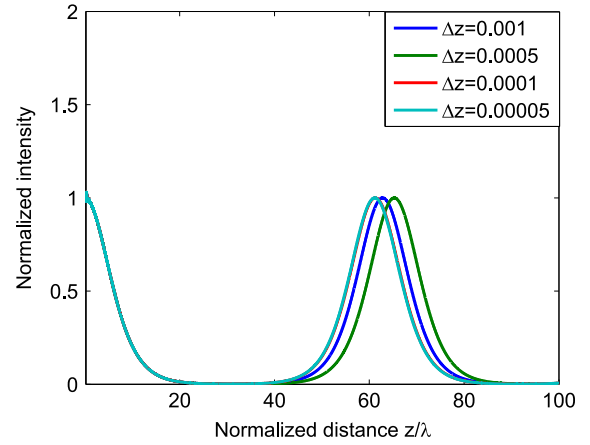


Fig. 1. Effect of Δz on envelope variations of the fundamental frequency. $\chi^{(1)}(2\omega_0) = \chi^{(1)}(\omega_0) = 2$, $\chi^{(2)}(2\omega_0) = \chi^{(2)}(\omega_0) = 2 \times 10^{-12}$ V/m, $E_{\omega_0}^0 = E_{\omega_0}^1 = 10 \times 10^8$ V/m and $E_{2\omega_0}^0 = E_{2\omega_0}^1 = 0$.

E_n^i and $E_{-n}^i = \bar{E}_n^i$. Assuming that a single plane-wave at frequency ω_0 enters a lossless and non-dispersive NL medium at $z=0$, Eqs. (11) and (12) are solved simultaneously to obtain the electric field at FF and SH. As in any other finite difference scheme, the choice of Δz strongly affects the accuracy and stability of the method. Fig. 1 shows envelope variations of the FF component for different values of Δz . For larger values of $\Delta z/\lambda$ the method is unstable and the envelope is not predicted correctly when compared to the available analytic solution in Fig. 2. However, as the step size is decreased, after some point the envelope remains almost unchanged which indicates the convergence of the method. For the set of parameters used in this paper, the results converge for $\Delta z < 0.0005\lambda$ and it was chosen to be $\Delta z = 0.0001\lambda$ in all simulations.

As the first example we apply the method to a non-dispersive weakly nonlinear medium in order to validate the numerical results by comparing them with those of the analytic solution. The medium susceptibilities are $\chi^{(1)}(\omega_0) = \chi^{(1)}(2\omega_0) = 3.48$ and $\chi^{(2)}(\omega_0) = \chi^{(2)}(2\omega_0) = 85 \times 10^{-12}$ V/m. The initial conditions are expressed by $E_{\omega_0}^0 = E_{\omega_0}^1 = 1.1 \times 10^8$ V/m and $E_{2\omega_0}^0 = E_{2\omega_0}^1 = 0$ which ensure the second harmonic does not enter the medium. The normalized field intensities are shown in Fig. 2. As the wave propagates in the medium, the SH is excited and its intensity increases while the intensity of the FF decreases. This behavior is also predicted by the analytical solution, which is shown in Fig. 2

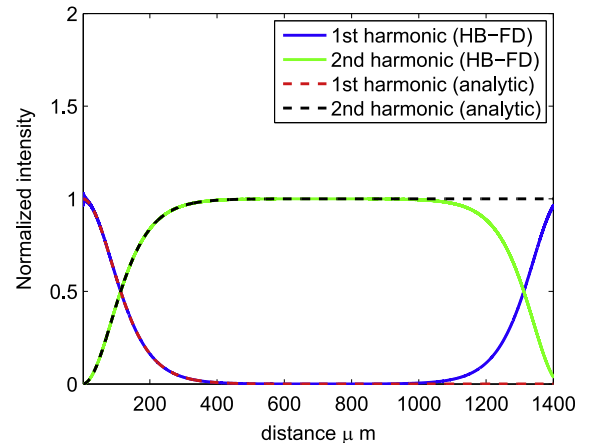


Fig. 2. Normalized intensities of the first and second harmonics in a nonlinear medium based on the HB-FD technique and analytic solution. $\chi^{(1)}(2\omega_0) = \chi^{(1)}(\omega_0) = 3.84$, $\chi^{(2)}(2\omega_0) = \chi^{(2)}(\omega_0) = 88 \times 10^{-12}$ V/m, $E_{\omega_0}^0 = E_{\omega_0}^1 = 1.1 \times 10^8$ V/m, $E_{2\omega_0}^0 = E_{2\omega_0}^1 = 0$, $\lambda_0 = 1.7776$ μm and $\Delta z' = 0.0001$.

by dashed lines [1,3,4], and also by other numerical solutions such as FDTD and BPM [28]. Evidently, the HB-FD results are in good agreement with the analytical solution up to $z \approx 1000 \mu\text{m}$. Eqs. (11) and (12) imply that the sub-harmonic frequency at ω_0 cannot be generated if a single plane-wave at frequency $2\omega_0$ enters the medium [3,29]. Consequently, if the intensity of FF reduces to zero at some point in the medium, it is expected to remain zero and the total input power to be transferred to the SH. However, as seen in Fig. 2, the intensity of FF does not diminish completely and starts to increase again while the intensity of SH starts to decay. Therefore, the input power is periodically exchanged between the FF and SH. Although this *regeneration* has also been predicted in [3] in a general solution of second harmonic generation, its period becomes infinite in the simplified analytical solution for phase matched condition. Extensive simulations reveal that the period of this exchange of power decreases if $\chi^{(2)}/\chi^{(1)}$ or the input power is increased and vice versa. In other words, this behavior is more pronounced when the nonlinearity is stronger. Fig. 3(a) and (b) demonstrates the above statement. In fact the input power is never totally transferred to the SH and complete disappearance of the FF predicted by the analytical solution is only the result of SVE approximation for weak nonlinearity. In fact, it can be shown that the second order derivative, which is neglected in SVE approximation, mathematically has an effect similar to phase mismatch in the analytical solution which leads to periodic envelope variation of FF and SH.

Based on Manley–Rowe relations the total power along the

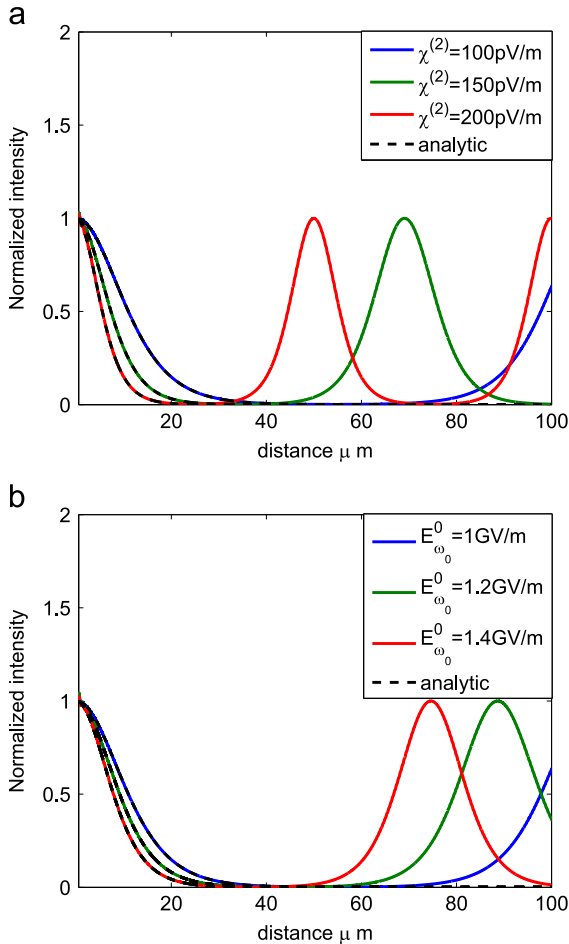


Fig. 3. Regeneration of the first harmonic for (a) different values of $\chi^{(2)}$ and (b) different values of input power. $\chi^{(1)}(2\omega_0) = \chi^{(1)}(\omega_0) = 3.84$, $E_{\omega_0}^0 = E_{2\omega_0}^0 = 0$, $\lambda_0 = 1.7776 \mu\text{m}$ and $\Delta z' = 0.0001$.

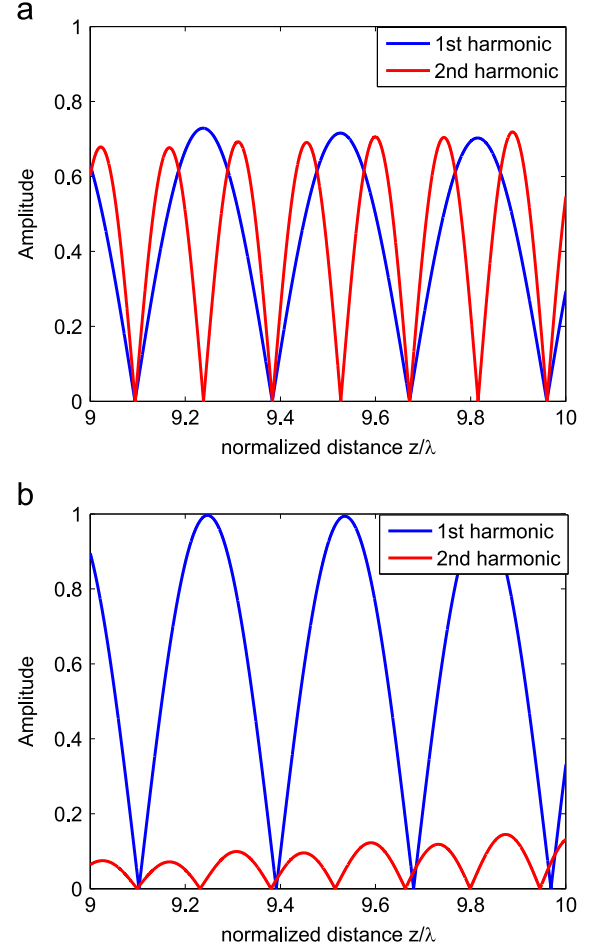


Fig. 4. Amplitude variations of the first and second harmonics in (a) non-dispersive and (b) dispersive media over one free-space wavelength. $\chi^{(2)}(2\omega_0) = \chi^{(2)}(\omega_0) = 50 \times 10^{-12} \text{ V/m}$, $E_{\omega_0}^0 = E_{\omega_0}^1 = 10 \times 10^8 \text{ V/m}$, $E_{2\omega_0}^0 = E_{2\omega_0}^1 = 0$ and $\Delta z' = 0.0001$.

propagation path in a lossless NL medium must remain constant [24,1]:

$$P_{\text{tot}} = \sum_{n=1}^{\infty} |A_n(z)|^2 = \text{constant} \quad (13)$$

In our example, we expect to have $|A_1(z)|^2 + |A_2(z)|^2 = |A_1(0)|^2 + |A_2(0)|^2 = 1$ which is evident from Fig. 2.

In a non-dispersive NL medium, it is expected that the two harmonics remain in-phase, i.e. $k_2 = 2k_1$. This behavior can be confirmed by studying the sinusoidal amplitude variations (not the envelopes) of the FF and SH within one free-space wavelength. Comparing the zero crossings confirms that $k_2 = 2k_1$.

In a dispersive medium, however, $\chi^{(1)}(2\omega_0) \neq \chi^{(1)}(\omega_0)$ and $2k_1 - k_2$ is nonzero. Thus, a phase mismatch builds up between the FF and SH [3]. An example of propagation in a dispersive medium is shown in Fig. 4. Dispersion also affects the envelope variations of each harmonic significantly. Fig. 5 shows envelope variations of the FF and SH for different values of $\chi^{(1)}(2\omega_0)/\chi^{(1)}(\omega_0)$. Their behavior shows that the effect of nonlinearity can be partly compensated by dispersion because as $\chi^{(1)}(2\omega_0)/\chi^{(1)}(\omega_0)$ increases the envelope of FF tends to remain almost constant throughout the medium. The envelope of SH varies sinusoidally as expected [3]. Its period of variations is commonly known as coherent length [30].

If the third harmonic (TH) is allowed to propagate in a NL

medium in which only $\chi^{(2)}$ is present and dominant, behavior of the FF and SH will also be affected significantly. No closed form analytical solution for this case has been presented before. However, the TH can be easily included in HB-FD method and the following equations are obtained from (9) after neglecting $\chi^{(3)}$ and keeping the harmonics up to third order:

$$\begin{aligned} E_{\omega_0}^{i+1} &= 2E_{\omega_0}^i - E_{\omega_0}^{i-1} \\ &- (2\pi\Delta z')^2 \left[(1 + \chi^{(1)}(\omega_0))E_{\omega_0}^i \right. \\ &+ 2\chi^{(2)}(\omega_0; 2\omega_0, -\omega_0)E_{2\omega_0}^i \bar{E}_{\omega_0}^i \\ &\left. + 2\chi^{(2)}(\omega_0; 3\omega_0, -2\omega_0)E_{3\omega_0}^i \bar{E}_{2\omega_0}^i \right] = 0 \end{aligned} \quad (14a)$$

$$\begin{aligned} E_{2\omega_0}^{i+1} &= 2E_{2\omega_0}^i - E_{2\omega_0}^{i-1} \\ &- (2\pi\Delta z')^2 \left[4(1 + \chi^{(1)}(2\omega_0))E_{2\omega_0}^i \right. \\ &+ 4\chi^{(2)}(2\omega_0; \omega_0, \omega_0)E_{\omega_0}^i E_{\omega_0}^i \\ &\left. + 8\chi^{(2)}(2\omega_0; 3\omega_0, -\omega_0)E_{3\omega_0}^i \bar{E}_{\omega_0}^i \right] = 0 \end{aligned} \quad (14b)$$

$$\begin{aligned} E_{3\omega_0}^{i+1} &= 2E_{3\omega_0}^i - E_{3\omega_0}^{i-1} \\ &- (2\pi\Delta z')^2 \left[9(1 + \chi^{(1)}(3\omega_0))E_{3\omega_0}^i \right. \\ &\left. + 18\chi^{(2)}(3\omega_0; \omega_0, 2\omega_0)E_{\omega_0}^i E_{2\omega_0}^i \right] = 0 \end{aligned} \quad (14c)$$

Numerical results for envelope variations are shown in Fig. 6 where it was assumed that a single plane-wave at frequency ω_0 enters a lossless NL medium at $z=0$. Note that both the SH and the TH are generated as the FF propagates through the medium. Again the Manley–Rowe relations are valid and the total power remains constant and equal to the input power. It is interesting to note that when the TH is allowed to propagate, a larger amount of the input power is transferred to the TH rather than the SH. Moreover, the envelope of FF does not completely vanish at any point. Fig. 7 shows the sinusoidal amplitude of the three waves over one period that reveals the phase matching between the FF and the higher harmonics, i.e. $k_3 = 3k_1$ and $k_2 = 2k_1$ because the medium was assumed to be dispersion-less.

3.2. Frequency mixing

When an electromagnetic wave with two distinct frequency components at ω_1 and ω_2 ($\omega_2 > \omega_1$) enters a NL medium with second order nonlinearity, new components at $\omega_2 \pm \omega_1$ are generated. This phenomenon is known as frequency mixing. In this case, after neglecting higher order frequency components, Eq. (9) is simplified to

$$\begin{aligned} E_1^{i+1} &= 2E_1^i - E_1^{i-1} - (2\pi\Delta z')^2 \left[(1 + \chi^{(1)}(\omega_1))E_1^i \right. \\ &+ 2\chi^{(2)}(\omega_1; \omega_1 + \omega_2, -\omega_2)E_3 \bar{E}_2 \\ &\left. + 2\chi^{(2)}(\omega_1; \omega_1 - \omega_2, \omega_2)E_4 \bar{E}_2 \right] \end{aligned} \quad (15a)$$

$$\begin{aligned} E_2^{i+1} &= 2E_2^i - E_2^{i-1} - (2\pi\Delta z')^2 \frac{\omega_2^2}{\omega_1^2} \left[(1 + \chi^{(1)}(\omega_2))E_2^i \right. \\ &+ 2\chi^{(2)}(\omega_2; \omega_1 + \omega_2, -\omega_1)E_3 \bar{E}_1 \\ &\left. + 2\chi^{(2)}(\omega_2; -(\omega_1 - \omega_2), \omega_1)E_1 \bar{E}_4 \right] \end{aligned} \quad (15b)$$

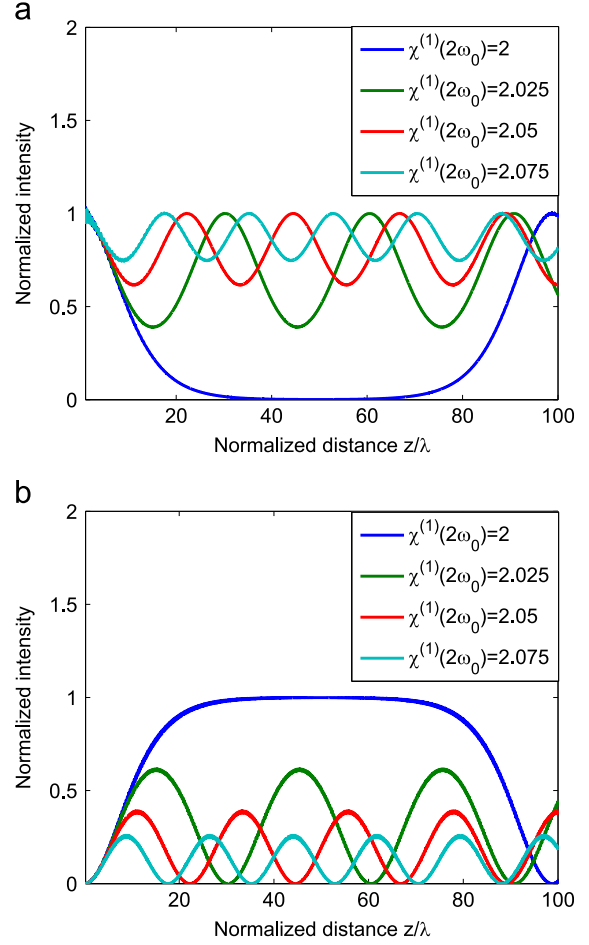


Fig. 5. Envelope variations of the (a) fundamental frequency (b) second harmonic in a dispersive medium for different values of $\chi^{(1)}(2\omega_0)$. $\chi^{(1)}(\omega_0) = 2$, $\chi^{(2)}(2\omega_0) = \chi^{(2)}(\omega_0) = 50 \times 10^{-12}$ V/m, $E_{\omega_0}^0 = E_{\omega_0}^1 = 10 \times 10^8$ V/m, $E_{2\omega_0}^0 = E_{2\omega_0}^1 = 0$ and $\Delta z' = 0.0001$.

$$\begin{aligned} E_3^{i+1} &= 2E_3^i - E_3^{i-1} - (2\pi\Delta z')^2 \left(1 + \frac{\omega_2}{\omega_1} \right)^2 \\ &\times \left[(1 + \chi^{(1)}(\omega_1 + \omega_2))E_3 + 2\chi^{(2)}(\omega_1 + \omega_2; \omega_1, \omega_2)E_1 E_2 \right] \end{aligned} \quad (15c)$$

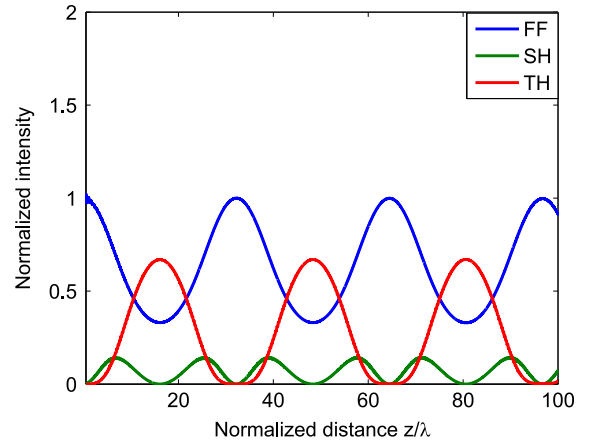


Fig. 6. Envelope variations of the first, second, and third harmonics in a NL medium. $\chi^{(1)}(3\omega_0) = \chi^{(1)}(2\omega_0) = \chi^{(1)}(\omega_0) = 2$, $\chi^{(2)}(3\omega_0) = \chi^{(2)}(2\omega_0) = \chi^{(2)}(\omega_0) = 50 \times 10^{-12}$ V/m, $E_{\omega_0}^0 = E_{\omega_0}^1 = 10 \times 10^8$ V/m, $E_{2\omega_0}^0 = E_{2\omega_0}^1 = 0$, $E_{3\omega_0}^0 = E_{3\omega_0}^1 = 0$ and $\Delta z' = 0.0001$.

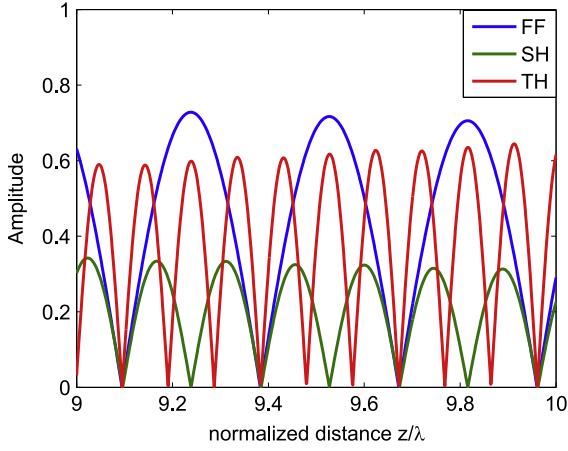


Fig. 7. Amplitude variations of the first, second, and third harmonics in a non-dispersive NL medium within one free-space wavelength. $\chi^{(1)}(3\omega_0) = \chi^{(1)}(2\omega_0) = \chi^{(1)}(\omega_0) = 2$, $\chi^{(2)}(3\omega_0) = \chi^{(2)}(2\omega_0) = \chi^{(2)}(\omega_0) = 50 \times 10^{-12}$ V/m, $E_{\omega_0}^0 = E_{\omega_0}^1 = 10 \times 10^8$ V/m, $E_{2\omega_0}^0 = E_{2\omega_0}^1 = 0$, $E_{3\omega_0}^0 = E_{3\omega_0}^1 = 0$ and $\Delta z' = 0.0001$.

$$E_4^{i+1} = 2E_4^i - E_4^{i-1} - (2\pi\Delta z')^2 \left(1 - \frac{\omega_2}{\omega_1}\right)^2 \times \left[(1 + \chi^{(1)}(\omega_1 - \omega_2))E_4 + 2\chi^{(2)}(\omega_1 - \omega_2; \omega_1, -\omega_2)E_1E_2 \right] \quad (15d)$$

where $\omega_1 = 2\pi c/\lambda_1$, $\Delta z' = \Delta z/\lambda_1$, and E_1, E_2, E_3, E_4 are the electric field components at $\omega_1, \omega_2, \omega_2 + \omega_1$, and $\omega_2 - \omega_1$, respectively. For numerical simulation, consider an electromagnetic wave with two components at ω_1 and $\omega_2 = 1.2\omega_1$ with the same amplitude and phase entering a non-dispersive NL medium at $z=0$. First, the component at $\omega_2 - \omega_1$ is ignored and only the sum frequency generation is simulated [1]. Envelope variations of the three frequency components ($\omega_1, \omega_2, \omega_2 + \omega_1$) are shown in Fig. 8. The Manly–Rowe relations are satisfied which validates the numerical results. The total power remains constant at all points:

$$P_{tot} = \sum_{n=1}^{n=3} |A_n(z)|^2 = 2 \quad (16)$$

Similarly, the difference frequency generation is simulated by neglecting the sum frequency component [1]. Fig. 9 shows the numerical results for the envelope variations. Note that the amplitude of the smaller fundamental frequency, ω_1 , rises above its initial value, i.e. it is amplified, and then decreases to its initial value before increasing again. This phenomenon is called optical parametric amplification which is an important result of difference frequency generation [1,29,30].

Furthermore, the sum and difference frequency generation could be considered simultaneously if both frequency components are allowed to propagate. The result, which is shown in Fig. 10, reveals that the parametric amplification does not occur in this case, i.e. in order to have parametric amplification we must stop the sum frequency from propagating. This behavior is not predicted by analytical solutions [1,29,30].

Propagation of second order harmonics, $2\omega_1$ and $2\omega_2$, together with the sum frequency component may be more practical than simultaneous propagation of sum and difference frequency components specially when ω_1 and ω_2 are very close to each other. Numerical results for $\omega_2 = 1.02\omega_1$ are shown in Fig. 11 which is obtained by considering $\omega_1, \omega_2, 2\omega_1, 2\omega_2, \omega_2 + \omega_1$ in (9). Also, it should be pointed out that the validity of Manly–Rowe relations is obviously clear as $P_{tot} = \sum_{n=1}^n |A_n(z)|^2 = 2$.

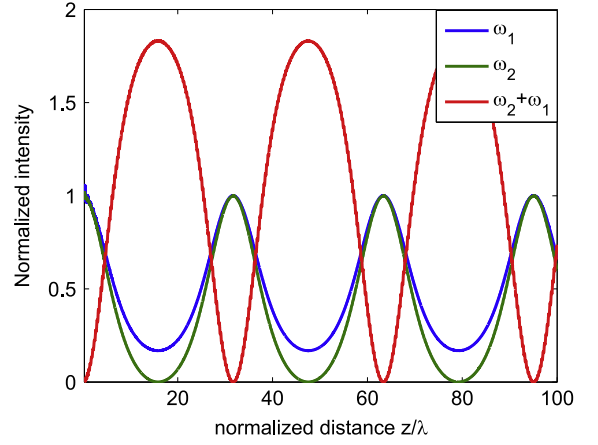


Fig. 8. The envelope variations of frequency components in sum frequency generation. $\omega_2 = 1.2\omega_1$, $\chi^{(1)}(\omega_2) = \chi^{(1)}(\omega_1) = 2$, $\chi^{(2)}(\omega_2) = \chi^{(2)}(\omega_1) = 50 \times 10^{-12}$ V/m, $E_{\omega_1}^0 = E_{\omega_1}^1 = 10 \times 10^8$ V/m, $E_{\omega_2}^0 = E_{\omega_2}^1 = 10 \times 10^8$ V/m.

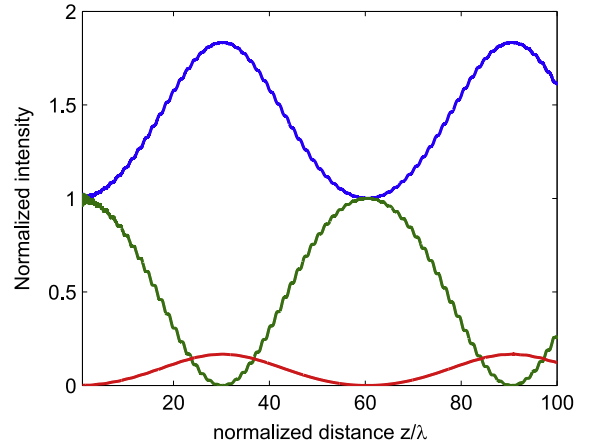


Fig. 9. The envelope variations of frequency components in difference frequency generation. $\omega_2 = 1.2\omega_1$, $\chi^{(1)}(\omega_2) = \chi^{(1)}(\omega_1) = 2$, $\chi^{(2)}(\omega_2) = \chi^{(2)}(\omega_1) = 50 \times 10^{-12}$ V/m, $E_{\omega_1}^0 = E_{\omega_1}^1 = 10 \times 10^8$ V/m, $E_{\omega_2}^0 = E_{\omega_2}^1 = 10 \times 10^8$ V/m.

4. Conclusion

A novel method, called HB-FD, was presented for complete

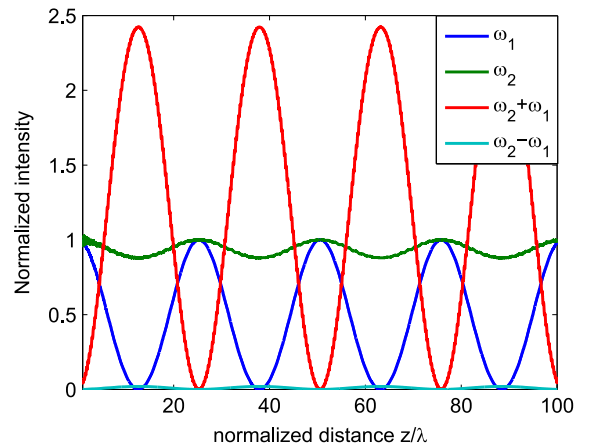


Fig. 10. The envelope variations of frequency components when both the sum and difference frequency generations are considered. $\omega_2 = 1.2\omega_1$, $\chi^{(2)}(\omega_2) = \chi^{(2)}(\omega_1) = 2$, $\chi^{(2)}(\omega_2) = \chi^{(2)}(\omega_1) = 50 \times 10^{-12}$ V/m, $E_{\omega_1}^0 = E_{\omega_1}^1 = 10 \times 10^8$ V/m, $E_{\omega_2}^0 = E_{\omega_2}^1 = 10 \times 10^8$ V/m.

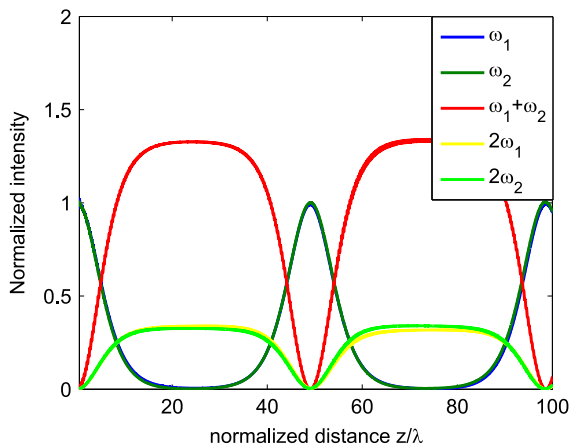


Fig. 11. The envelope variations of frequency components when second harmonics and sum frequency are considered simultaneously. $\omega_2 = 1.02\omega_1$, $\chi^{(2)}(\omega_2) = \chi^{(2)}(\omega_1) = 2$, $\chi^{(2)}(\omega_2) = \chi^{(2)}(\omega_1) = 50 \times 10^{-12} \text{V/m}$, $E_{\omega_1}^0 = E_{\omega_1}^1 = 10 \times 10^8 \text{V/m}$, $E_{\omega_2}^0 = E_{\omega_2}^1 = 10 \times 10^8 \text{V/m}$.

steady state time-harmonic solution of the nonlinear wave equation which combines the harmonic balance and finite difference methods. All interactions between the fundamental and higher order harmonics, up to an arbitrary order, can be explicitly incorporated into the solution. Considering plane-wave propagation in a homogeneous nonlinear medium, several nonlinear phenomena including SHG and frequency mixing were simulated with the new technique to verify its accuracy and versatility. The results are in good agreement with analytical solutions whenever they are available. In more complex situations, where a larger number of frequency components are involved, no analytical solution is available and numerical results were verified by checking the power conservation relation. Since susceptibilities can be complex valued and inhomogeneous, many other phenomena such as Backward SHG [31] can also be studied with HB-FD method. Major advantages of the proposed method over the available numerical and analytical techniques are: (i) capability of considering several harmonics or several waves at different frequencies propagating together, (ii) the envelope of the wave is not assumed to be slowly varying, (iii) true dispersion characteristics of the medium are correctly taken into account.

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