High-order optical processes in intense laser field: Towards nonperturbative nonlinear optics

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We develop an approach describing nonlinear-optical processes in the strong-field domain characterized by the nonperturbative field-with-matter interaction. The polarization of an isolated atom in the external field calculated via the numerical solution of the time-dependent Schrödinger equation agrees with our analytical findings. For the practically important case of one strong laser field and several weaker fields, we derive and analytically solve propagation equations describing high-order (HO) wave mixing, HO parametric amplification, and HO stimulated scattering. These processes provide a way of efficient coherent xuv generation. Some properties of HO processes are new in nonlinear optics: essentially complex values of the coefficients in the propagation equations, the superexponential (hyperbolic) growing solutions, etc. Finally, we suggest conditions for the practical realization of these processes and discuss published numerical and experimental results where such processes could have been observed.

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I. INTRODUCTION

Nonlinear optics usually deals with the laser field which perturbatively interacts with matter. In this case, the matter response is described with nonlinear susceptibilities $\chi^{(m)}$. This approach allows one to investigate numerous nonlinear-optical effects involving few photons [1]. However, in the case of intense laser field, the perturbation approach fails. A number of multiphoton processes involving *electronic* dynamics (ionization, electronic rescattering, etc.) are successfully described within nonperturbative approaches (such as Keldysh approximation [2]), but the only fully *optical* process which is well understood in this case is high-order harmonic generation (HHG). The study of the other nonlinear optical processes in the nonperturbative regime is limited, among other factors, by poorly developed theoretical methods of their description. For instance, experimental [3] observation of the exponential growth of the high harmonic signal has led to an active discussion [4–6]. The feasibility of high-order optical processes was already discussed in the early studies [7–10], but the theoretical methods were based on using nonlinear susceptibilities and thus were limited to processes of relatively low order. More recently, the xuv amplification was obtained in simulations [11] based on direct numerical integration of the propagation equation with the nonlinear polarization calculated via numerical solution of the time-dependent Schrödinger equation (TDSE).

In this paper, we suggest an approach to describe nonlinear optical processes in the presence of a given strong laser field (denoted as E_0). Processes involving other fields (denoted as $E_{1,2,\ldots}$) are described with the susceptibilities, nonperturbatively induced by the pump field. The remarkable practical importance of the suggested approach is that even in the strong-field domain, we write and in several cases solve analytically the propagation equations for the fields $E_{1,2,\ldots}$. To study the conditions for which such processes can be observed experimentally, we calculate the induced susceptibilities for a model Xe atom using a numerical TDSE solution. Our results show that efficient coherent xuv generation can be one of the applications of the HO nonlinear processes.

II. EXPANSION OF THE MICROSCOPIC NONLINEAR POLARIZATION

Let us consider the microscopic polarization introduced by two external fields E_0 and E_1. Irrespective of the nature of nonlinearity, the microscopic polarization $P(t)$ is a *functional* of the external field $E = E_0 + E_1$,

$$P(t) = \Phi[E] = \Phi[E_0 + E_1].$$

The functional can be expanded in the Taylor series [12] (for some physical applications of such expansion, see [13,14]):

$$P(t) = P^{(0)}(t) + P^{(1)}(t) + P^{(2)}(t) + \cdots ,$$

where

$$P^{(0)}(t) = \Phi[E_0], \tag{1}$$

$$P^{(1)}(t) = \int_0^{+\infty} d\tau \left.\frac{\delta\Phi}{\delta E}\right|_{E=E_0(t-\tau)} E_1(t-\tau) + \text{c.c.}, \tag{2}$$

$$P^{(2)}(t) = \int_0^{+\infty} d\tau \int_0^{+\infty} d\tau' \left.\frac{1}{2}\frac{\delta^2\Phi}{\delta E^2}\right|_{E=E_0(t-\tau')} \times E_1(t-\tau)E_1(t-\tau') + \text{c.c.}, \tag{3}$$

where $\frac{\delta\Phi}{\delta E}$, $\frac{\delta^2\Phi}{\delta E^2}$ are the *functional* derivatives of the functional Φ over the function E.

Let us consider fields

$$E_{0,1} = \mathcal{E}_{0,1} \exp(-i\omega_{0,1} t) + \text{c.c.}$$

Periodicity of the field E_0 allows the expansion of the functional derivatives in Eqs. (2) and (3) in the Fourier

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series,

$$\left.\frac{\delta\Phi}{\delta E}\right|_{E=E_0(t-\tau)} = \sum_q G_q^{(1)}(\mathcal{E}_0,\omega_0,\tau)\exp[-iq\omega_0(t-\tau)], \quad (4)$$

$$\left.\frac{1}{2}\frac{\delta^2\Phi}{\delta E^2}\right|_{E=E_0(t-\tau')} = \sum_q G_q^{(2)}(\mathcal{E}_0,\omega_0,\tau')\exp[-iq\omega_0(t-\tau')]. \quad (5)$$

Substituting these expansions into Eqs. (2) and (3), we have

$$P^{(1)}(t) = \sum_q \mathcal{E}_1 \exp[-i(q\omega_0+\omega_1)t]\kappa_q^{(+1)} + \mathcal{E}_1^* \exp[-i(q\omega_0-\omega_1)t]\kappa_q^{(-1)} + \text{c.c.}, \quad (6)$$

$$P^{(2)}(t) = \sum_q \mathcal{E}_1^2 \exp[-i(q\omega_0+2\omega_1)t]\kappa_q^{(+2)} + (\mathcal{E}_1^*)^2 \exp[-i(q\omega_0-2\omega_1)t]\kappa_q^{(-2)} + |\mathcal{E}_1|^2 \exp(-iq\omega_0 t)\kappa_q^{(0,2)} + \text{c.c.}, \quad (7)$$

where

$$\kappa_q^{(\pm1)}(\mathcal{E}_0,\omega_0,\omega_1) = \int_0^{+\infty} d\tau G_q^{(1)}(\mathcal{E}_0,\omega_0,\tau) \times \exp(iq\omega_0\tau \pm i\omega_1\tau), \quad (8)$$

$$\kappa_q^{(\pm2)}(\mathcal{E}_0,\omega_0,\omega_1) = \int_0^{+\infty} d\tau \int_0^{+\infty} d\tau' G_q^{(2)}(\mathcal{E}_0,\omega_0,\tau') \times \exp[iq\omega_0\tau' \pm i\omega_1(\tau+\tau')], \quad (9)$$

$$\kappa_q^{(0,2)}(\mathcal{E}_0,\omega_0,\omega_1) = \int_0^{+\infty} d\tau \int_0^{+\infty} d\tau' G_q^{(2)}(\mathcal{E}_0,\omega_0,\tau') \times \exp(iq\omega_0\tau')\{\exp[i\omega_1(\tau-\tau')] + \exp[-i\omega_1(\tau-\tau')]\}. \quad (10)$$

Under certain conditions (see [15] and references therein), the functional $\Phi[E_0]$ in Eq. (1) can be expanded in Fourier series according to the Floquet theorem,

$$P^{(0)}(t) = \sum_q \kappa_q^{(0)}(\mathcal{E}_0,\omega_0)\exp(-iq\omega_0 t) + \text{c.c.} \quad (11)$$

Thus the Fourier expansion of the microscopic response P can be written as

$$P(t) = \sum_{q=-\infty}^{+\infty}\sum_{m=-\infty}^{+\infty} \tilde{\kappa}_q^{(m)}(\mathcal{E}_1)^{|m|}\exp\{-iq\omega_0 t - im\omega_1 t\} + \text{c.c.}, \quad (12)$$

where

$$\tilde{\kappa}_q^{(m)}(\mathcal{E}_0,\mathcal{E}_1,\omega_0,\omega_1) = \sum_{j=0,2,\ldots}^{+\infty} \kappa_q^{(m,j)}(\mathcal{E}_0,\omega_0,\omega_1)|\mathcal{E}_1|^j, \quad (13)$$

$$\kappa_q^{(m,0)} \equiv \kappa_q^{(m)}.$$

From Eq. (13), one can see that terms with $j = 2,4,\ldots$ can be understood as those describing processes where one, two, etc. photons from the field E_1 were absorbed and the same number of photons of this field were emitted.

Now let us consider the fields

$$E_{0,1}(\mathbf{r},t) = \mathcal{E}_{0,1}\exp(-i\omega_{0,1}t + \mathbf{k}_{0,1}\mathbf{r} + \varphi_{0,1}) + \text{c.c.}, \quad (14)$$

with the real amplitudes $\mathcal{E}_{0,1}$.

Assuming the locality of the microscopic response and substituting $-\omega_{0,1}t \to -\omega_{0,1}t + \mathbf{k}_{0,1}\mathbf{r} + \varphi_{0,1}$ in Eq. (12), we have

$$P(\mathbf{r},t) = \sum_{q=-\infty}^{+\infty}\sum_{m=-\infty}^{+\infty} \tilde{\kappa}_q^{(m)}(\mathcal{E}_1)^{|m|}\exp\{iq(-\omega_0 t + \mathbf{k}_0\mathbf{r} + \varphi_0) + im(-\omega_1 t + \mathbf{k}_1\mathbf{r} + \varphi_1)\} + \text{c.c.}, \quad (15)$$

where the induced susceptibilities $\tilde{\kappa}_q^{(m)}$ are given by Eq. (13).

Thus, the microscopic response is a sum of waves to which q photons from the one field and $|m| + j$ photons from the other one contribute. For the case $\omega_0 = \omega_1$, $\mathbf{k}_0 \neq \mathbf{k}_1$, a similar equation was found in Ref. [16]. Note that up to here we never supposed that the field $\mathcal{E}_1$ is small. Terms with $j = 2,4,\ldots$ in Eq. (13) can be understood as those describing processes where one, two, etc. photons from the field E_1 were absorbed and the same number of photons of this field were emitted.

The remarkable feature of Eq. (15) is the fact that κ depends only on the real amplitudes of the fields, and the dependence on the fields' phases is only in the exponent. At a first glance, this contradicts the fact that for the HHG in the two-color field consisting of the fundamental and its second harmonic, the high harmonic amplitude and phase depend on the phase difference of the generating fields in a complex way [17–19]. Note, however, that when ω_0 and ω_1 are multiple numbers (for instance, for $\omega_1 = 2\omega_0$), different q and m can provide contributions with the same frequency; the interference of these terms provides the complex dependence on the dephasing. Below we consider only the case of linearly polarized fields. However, for the case of two-color circularly polarized fields [20–22], a corresponding generalization of our approach could be useful. Moreover, the comparison with the description of the nonlinear-optical effects in the strong-field region suggested in [21,22] can be done.

When $\mathcal{E}_1 \ll \mathcal{E}_0$, the first term in Eq. (13) dominates, so we have $\tilde{\kappa}_q^{(m)}(\mathcal{E}_0,\mathcal{E}_1,\omega_0,\omega_1) \approx \kappa_q^{(m,0)}(\mathcal{E}_0,\omega_0,\omega_1)$. As $\kappa_q^{(m,0)}$ does not depend on $\mathcal{E}_1$, in the expansion (15) the dependence on the field $\mathcal{E}_1$ remains *only* in the term $(\mathcal{E}_1)^{|m|}$. This makes this expansion for the polarization similar to the one appearing in the usual ("perturbative") nonlinear optics [1]. Below, for brevity, we write $\kappa_q^{(m)} \equiv \kappa_q^{(m,0)}$.

Note that in the case of absence of the pump field, Eq. (15) turns into the well-known perturbative expansion of the polarization [1]:

$$P^{\text{per}}(t) = \sum_{m=1}^{+\infty} \chi^{(m)}(\mathcal{E}_1)^m \exp\{im(-\omega_1 t + \mathbf{k}_1\mathbf{r} + \varphi_1)\} + \text{c.c.}$$

As we mentioned in Sec. I, the practical importance of Eq. (15) is that even in the strong-field domain, one can write and sometimes solve analytically the propagation equation with the right side given by Eq. (15). So there is no need to calculate the nonperturbative response simultaneously with the propagation equation solution. Instead, one should find the polarization using an appropriate theoretical approach (this can be the numerical TDSE solution, strong-field

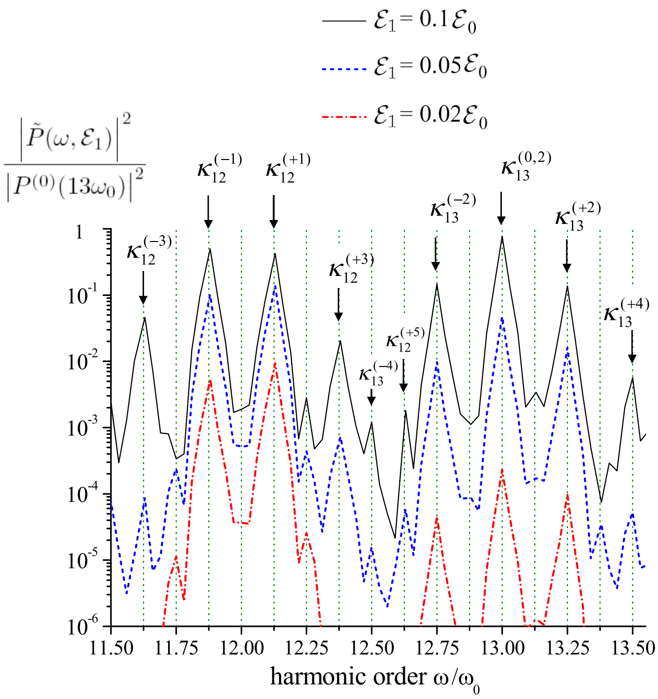

FIG. 1. Spectra of the polarization induced by the weak field E_1 in the presence of the intense field E_0 for different strengths of the weak field; the spectra are calculated numerically via TDSE solution for a model Xe atom (see text for more details). Arrows show the spectral components generated due to different induced susceptibilities $\kappa_q^{(m)}$. The spectra are normalized with the intensity of the 13th harmonic generated in the absence of the weak field.

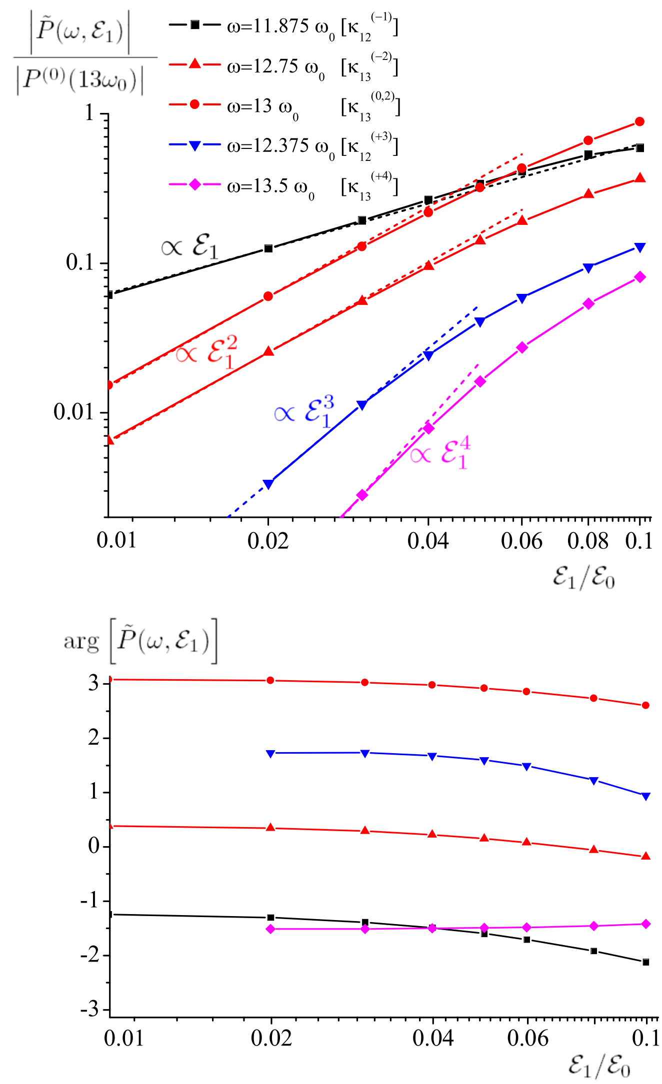

FIG. 2. Absolute values (upper panel) and phases (lower panel) of spectral components of the response shown in Fig. 1 as a function of the weak field strength. Several components with different frequencies are shown; the induced susceptibility responsible for the generation of every component is presented in square brackets. Dotted lines show the power approximations.

approximation [16,23,24], or classical models [3,25] for an isolated atom response, particle-in-cell simulations for plasma, etc.), calculate its spectrum $P(\omega)$, and then find κ from this spectrum using Eq. (15); alternatively, one can directly calculate κ from Eqs. (8)–(10).

To check our analytical findings, we compare the found properties of the polarization with numerical results. We solve numerically the three-dimensional (3D) TDSE in the single-active electron approximation for a model Xe atom in an external laser field; the details of the TDSE solution are presented in Refs. [26,27]. For the given strong field E_0, we calculate numerically the polarization P in the presence of the weak field E_1 and the polarization $P^{(0)}$ in the absence of this field. Figure 1 presents the spectrum $\tilde{P}(\omega,\mathcal{E}_1) = P(\omega,\mathcal{E}_1) - P^{(0)}(\omega)$ for the given $\mathcal{E}_0$ and different $\mathcal{E}_1$. The strong-field wavelength is 800 nm and its intensity is 5×10^{13} W/cm^2. The weak field has low frequency $\omega_1 = \frac{1}{8}\omega_0$ and low intensity, which differs in different calculations.

We can see that in agreement with the selection rule for a central-inverse medium, only processes with an odd number of photons $q + |m| + j$ contribute to the process (for $|m| \leqslant 2$ and $j = 0$, this selection rule was demonstrated already in the early experiment [28]). The strongest contributions are the "doublet" around the even harmonic of the pump field $q = 12$ due to the $|m| = 1$, $j = 0$ processes [given by Eq. (6)], and the "triplet" around the odd one $q = 13$ due to the $|m| + j = 2$ processes [given by Eq. (7)].

Figure 2 shows the absolute value and the phase of the response $\tilde{P}(\omega,\mathcal{E}_1)$ as a function of the field $\mathcal{E}_1$. We can see that in agreement with our analytical findings, the arguments are approximately constant and the absolute values increase approximately as $\mathcal{E}_1^{|m|+j}$, with lowest possible j. However, the deviation from these (weak $\mathcal{E}_1$ field) approximations increases with $\mathcal{E}_1$ because terms with higher j should be taken into account in the expansions (13) and (15). The role of terms with $j \neq 0$ can be seen in this figure from the component with $\omega = 13\omega_0$ (red line with circles) generated due to $\kappa_{13}^{(0,2)}$. For $\mathcal{E}_1 = 0.1\mathcal{E}_0$, we can see that $|\tilde{P}(13\omega_0)| \approx |P^{(0)}(13\omega_0)|$, so this contribution is not negligible for such $\mathcal{E}_1$. Below we deal with lower fields $\mathcal{E}_1$, thus studying only the processes with $j = 0$.

Note that if we are really in the nonperturbative domain (i.e., $\mathcal{E}_1 \ll \mathcal{E}_0$ is not valid), $\tilde{\kappa}$ should be calculated (for instance, with the strong-field approximation or using numerical

TDSE solution), and the propagation equation should be integrated taking into account the dependence of $\tilde{\kappa}$ on the real amplitude $\mathcal{E}_1$.

III. SOME PROPERTIES OF THE INDUCED SUSCEPTIBILITIES

Below we consider only the centrally symmetric case (the medium is centrally symmetric and the fields are linearly polarized in the same plane). The following properties of the induced susceptibilities are found:

(i) From the numerical TDSE solution, we find that while q is in the plateau region, $\kappa_q^{(m)}$ are comparable for different orders q.

(ii) For $\omega_1 \ll q\omega_0$ and in the absence of resonances from Eq. (8), one can see that

$$\kappa_q^{(1)}(\omega_1) \approx \kappa_q^{(-1)}(\omega_1). \tag{16}$$

From Eq. (9) for $\kappa_q^{(2)}$ and from similar equations for higher-order induced susceptibilities, we have, under $m\omega_1 \ll q\omega_0$,

$$\kappa_q^{(m)}(\omega_1) \approx \kappa_q^{(-m)}(\omega_1). \tag{17}$$

For $m = 1,2$, this can be seen from Fig. 1. Moreover, for higher m, the difference between $\kappa_q^{(m)}$ and $\kappa_q^{(-m)}$ is visible.

(iii) In the following studies, we shall write explicitly the frequency of the response, using the following notations:

$$\kappa^{(1)}(\omega = q\omega_0 \pm \omega_1) \equiv \kappa_q^{(\pm 1)}(\omega_1),$$

$$\kappa^{(2)}(\omega = q\omega_0 \pm 2\omega_1) \equiv \kappa_q^{(\pm 2)}(\omega_1).$$

The permutation symmetry of the second-order nonlinear susceptibilities is [1]

$$\chi^{(2)*}(\omega = \omega_1 + \omega_2) = \chi^{(2)}(\omega_1 = -\omega_2 + \omega) = \chi^{(2)}(\omega_2 = \omega - \omega_1). \tag{18}$$

In the approximation of the given pump field, the first equation does not have its analog for the induced susceptibilities. However, the second equation has the analog:

$$|\kappa^{(1)}(\omega_2 = q\omega_0 - \omega_1)| = |\kappa^{(1)}(\omega_1 = -\omega_2 + q\omega_0)|. \tag{19}$$

Note that the equation takes place only for the absolute values. We shall derive this equation in Appendix A. It can also be derived directly from the Manley-Rowe relations, as is done in [29] for χ.

IV. SOME HIGH-ORDER NONLINEAR PROCESSES

Below we study several high-order nonlinear processes, illustrated in Fig. 3.

A. Wave mixing

The wave mixing is the process where q photons from the intense field and m photons from the weaker field are converted in one $\omega_2 = q\omega_0 + m\omega_1$ photon. In the plane-wave approximation and slowly varying amplitude approximation, directing the z axis along $q\mathbf{k}_0 + m\mathbf{k}_1$, we present the generated field as

$$E_2 = \mathcal{E}_2(z)\exp\{i(k_2 z - \omega_2 t)\}. \tag{20}$$

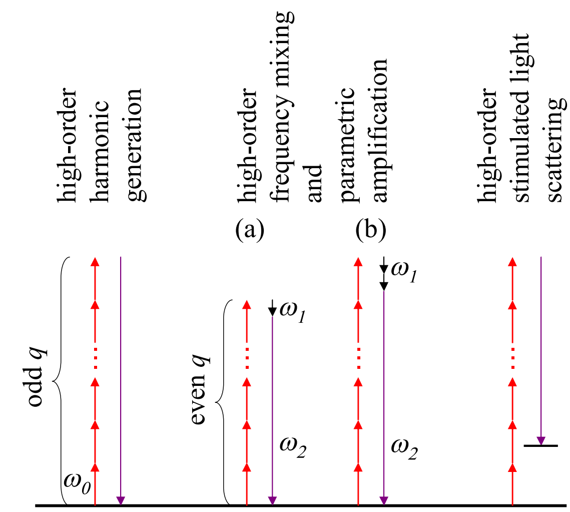

FIG. 3. The schematic of several high-order processes involving many photons from the pump field with frequency ω_0 and few photons from the weaker fields with frequencies ω_1 and ω_2.

The complex amplitude of the field $\mathcal{E}_2$ is described with the following propagation equation:

$$\frac{\partial \mathcal{E}_2}{\partial z} = \frac{i2\pi\omega_2^2}{k_2 c^2} P^{NL}(\omega_2, z)\exp\{-i(k_2 z - \omega_2 t)\}, \tag{21}$$

where P^{NL} is the nonlinear part of the polarization given by Eq. (15). From Eqs. (21) and (15), one can see that the intensity of the generated field $\mathcal{E}_2$ is proportional to the mth power of the intensity of the field $\mathcal{E}_1$. This explains the experimental findings of [30], where for xuv generation this behavior was observed up to $m = 6$. Another example is generation of THz radiation using a frequency-tunable half harmonic of a femtosecond pulse [31]. This process can be understood as difference-frequency generation (one fundamental photon plus $m = -2$ half harmonic photons). The experimentally found dependence of the THz yield on the fundamental intensity is much stronger than linear (as it should be in the intense-field domain), but the dependence on the weak half harmonic intensity is quadratic, as it should be for the process with $|m| = 2$.

From Eqs. (21) and (15), one can see that the detuning from the phase matching for a process of mixing of q photons from one beam and m photons from the other is, irrespective of the intensity ratio,

$$\Delta\mathbf{k}_{q,m} = q\mathbf{k}_0 + m\mathbf{k}_1 - \mathbf{k}_2, \tag{22}$$

where k_2 is the wave vector at the frequency ω_2. In Ref. [16], the phase matching for the noncollinear HHG ($\omega_1 = \omega_0$, $\mathbf{k}_1 \neq \mathbf{k}_0$) was considered. Here we assume $\omega_1 \neq \omega_0$, codirected $\mathbf{k}_1$ and $\mathbf{k}_0$, and plasma and/or capillary contribution to the dispersion dominates. In this case, the refraction index for some frequency $\tilde{\omega} \gg \omega_{pl}$ is $n = 1 - \omega_{pl}^2/(2\tilde{\omega}^2)$. Let us assume that the plasma frequency $\omega_{pl} \ll \omega_0, \omega_1$. Then,

$$\Delta k_{q,m} = \frac{\omega_{pl}^2}{2c}\left(-\frac{q}{\omega_0} - \frac{m}{\omega_1} + \frac{1}{q\omega_0 + m\omega_1}\right).$$

Omitting the last term, we find that for $m = -\frac{\omega_1}{\omega_2}$, the detuning is zero *irrespective of the plasma frequency*, and thus irrespective of its density. This is very important because the medium ionization always accompanies processes in a strong laser field and thus the plasma density is time varying. Note that both q and m are integer numbers and the total number of the involved photons $|q| + |m|$ should be odd.

The possibility of the phase-matching optimization in the difference-frequency mixing in plasma, involving few photons from the two waves, was first shown in Refs. [32,33] and further studied in Ref. [34]. For the case $\omega_0 = 2\omega_1$, $q = 6$, $m = -3$, the phase matching was experimentally demonstrated in Ref. [35]. However, the case of $\omega_0 = 2\omega_1$ is hardly perspective for solving the phase-matching problem for really high harmonics because, along with the optimal ($m = -q/2$) polarization wave, many other polarization waves are generated. This is not the case when only a few waves with different (small) m are generated. For $|m| \ll |q|$, it should be $\omega_1 \ll \omega_0$, and that is why in Figs. 1 and 2 we present the numerical results for this case. For $\omega_1 = \frac{1}{8}\omega_0$, the phase matching is achieved for $q = 8$ and $m = -1$, $q = 24$ and $m = -3$, and so on. In general, to achieve phase-matched generation of xuv using given number q and lowest $|m|$, for the case of even q one should use frequency $\omega_1 = \omega_0/q$ and $m = -1$ process, and for odd q one should use frequency $\omega_1 = 2\omega_0/q$ and $m = -2$ process; see Fig. 3 (see also the "duplet" near the even harmonic and the "triplet" near the odd one in Fig. 1). Both of these cases will be considered below in more detail.

B. Parametric amplification and generation

The process of the parametric amplification is described with the same equations as the difference-frequency generation. The difference is in the input conditions: the former assumes that initially there is one intense pump field and both generated fields are initially weak, while the latter assumes two initial intense fields [1].

1. Generation of two photons

Let us consider the process in which q photons from the initial field are converted into one ω_1 photon and one ω_2 photon; see Fig. 3(a). In this case, q is an even number (note that for $q = 2$, this is a well-known process of four-wave mixing; however, for higher q, the analogy with the four-wave mixing is hardly helpful). For $m = -1$, we have, from Eq. (15),

$$P^{NL}(\omega_{1,2},z) = \kappa_q^{(1)}(\omega_{1,2} = q\omega_0 - \omega_{2,1}) \times \exp\{iq(k_0 z - \omega_0 t + \varphi_0)\}E_{2,1}^*. \quad (23)$$

Substituting this polarization into the propagation equation, we have

$$\partial\mathcal{E}_{1,2}/\partial z = i2\pi k_{1,2}\kappa_q^{(1)}(\omega_{1,2})\mathcal{E}_{2,1}^* \exp\{i\Delta k z + iq\varphi_0\}, \quad (24)$$

where Δk given by Eq. (22) in the considered case is written as $\Delta k = qk_0 - k_1 - k_2$.

These equations are similar to those describing the parametric amplification in the perturbative nonlinear optics (see [1], part 9.1). In particular, the phase matching plays a critical role in this process defining the frequencies $\omega_{1,2}$ (and, in the case of the HO process, also the order q) which are generated with significant efficiencies. However, an important difference from the case of the perturbative nonlinear optics is that the susceptibility $\kappa_q^{(m)}$ is complex and its phase is not negligible. For instance, the phase of $\kappa_q^{(0)}$ is well understood within the recollision picture (see [36] and references therein). The details of the solution of the propagation equations (24) are presented in Appendix A. For the phase-matched process, we obtain the solution in which the exponential growth,

$$\mathcal{E}_{1,2}(z) \propto \exp\{g^{\pm}z\},$$

where $g^{\pm} = 2\pi\sqrt{k_1 k_2}|\kappa_q^{(1)}|\exp i\psi^{\pm}$, $\alpha = \sqrt{\omega_1/\omega_2}$, $\psi^+ = -\Delta\theta/2$, $\psi^- = -\Delta\theta/2 + \pi$, and $\Delta\theta = \arg[\kappa_q^{(1)}(\omega_2)] - \arg[\kappa_q^{(1)}(\omega_1)]$, dominates after a certain propagation distance.

To study the conditions for which the found exponential growth can be observed experimentally, we consider below a specific example. Namely, $\kappa_q^{(1)}$ is found for Xe in the pump laser field with intensity of 5×10^{13} W/cm^2 from the numerical TDSE solution described above as the limit of $P(q\omega_0 - \omega_1,\mathcal{E}_1)/\mathcal{E}_1$ for weak $\mathcal{E}_1$:

$$\kappa_q^{(1)} = \lim_{\mathcal{E}_1 \to 0} P(q\omega_0 - \omega_1,\mathcal{E}_1)/\mathcal{E}_1. \quad (25)$$

Our numerical calculations show that within the plateau region, $|\kappa_q^{(1)}|$ is almost independent of q. For the conditions of Fig. 1, we find that for $q = 12$, $|\kappa_q^{(1)}| = 4.5 \times 10^{-26} n_{\rm gas}$ CGS units, where $n_{\rm gas}$ is the gas density (in cm^{-3}). According to Eq. (A12) (see Appendix A), this gives the exponential growth of the intensity with the increment $g = 1.35$ cm^{-1} for the atmospheric pressure. Figure 4(a) shows the fields' intensities calculated for these conditions, under zero incident intensity of the first field. One can see that initially ($z < 1$ cm) the intensity of the first field grows quadratically, so the solution describes difference-frequency generation, and for $z > 2$ cm it grows exponentially. Note that the solution in the transition region can be less smooth depending on the initial phases of the fields and phases of the induced susceptibilities.

The found values of increment g show that the high-order parametric process can hardly be observed in atomic gas jets, but it can be observed in larger targets (capillaries and cells) especially under high gas pressure (pressures up to tens of atmospheres were used in recent experiments [37]). Moreover, this process can be very efficient in a parametric generator.

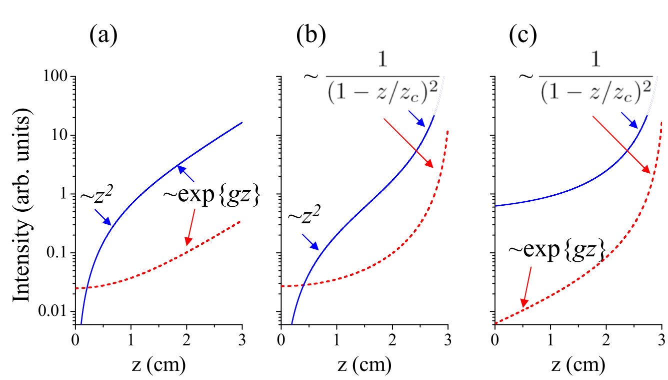

FIG. 4. Intensities of the fields E_1 (dotted red line) and E_2 (solid blue line) as a function of the propagation length for (a) two-photon parametric amplification and (b),(c) three-photon parametric amplification for different initial intensities of the fields.

Such parametric generator can be only a singly resonant one (i.e., the mirrors reflect the pump laser field E_0 and the weak low-frequency field E_1) because of the extremely low efficiency of optical elements in xuv. Note that the intracavity HHG was demonstrated experimentally [38] and now it is actively used [39,40].

Otherwise, the high-order parametric generation process can provide two photons with *comparable* frequencies. The phase-matching problem can be solved in noncollinear geometry. The other way is to utilize dispersion properties near some resonance: when one of the generated frequencies is close to the resonance, the phase matching can be achieved and the parametric generation process would be effective for these frequencies. For both cases, it is important that the plasma density is approximately constant for a given "retarded time" $\xi = t - z/c$, so the amplification for a given ξ takes place approximately as in the stationary case described above; however, the conditions providing phase matching (the angle in the first case or the detuning from the resonance in the second one) should be different for different ξ.

2. Generation of three photons

Another parametric process, which was not considered in the perturbative nonlinear optics, is the one where q photons from the pump field are split into more than two photons. Here we shall study the process where there are two low-frequency photons (ω_1) and one xuv photon (ω_2). Here, q should be odd, so under $\omega_1 \ll \omega_0$ the generated xuv frequency ω_2 is close to the qth harmonic frequency. Thus, the naturally broadened HH line can provide an effective seeding for this process, and this determines its practical importance. In Appendix B, we obtain the following propagation equations describing this process:

$$\partial\mathcal{E}_1/\partial z = A_1\mathcal{E}_1^*\mathcal{E}_2^* \exp\{i\Delta kz + iq\varphi_0\},$$
$$\partial\mathcal{E}_2/\partial z = A_2\left(\mathcal{E}_1^*\right)^2 \exp\{i\Delta kz + iq\varphi_0\}, \quad (26)$$

where $\Delta k = qk_0 - 2k_1 - k_2$. We solve it analytically for $\Delta k = 0$. The main property of the found analytical solution is the *hyperbolic* growth after a certain propagation distance, $\mathcal{E}_{1,2}(z) \propto 1/(1 - z/z_c)$, where z_c is the propagation distance characterizing a *catastrophe* (this is a standard term used in the Riccati equation solution; see Appendix B). Certainly, when z is close to z_c, the approximation of the given pump field fails, but the efficiency of the process should be high for such z.

Figure 4 shows the fields' intensities calculated for zero incident first field [Fig. 4(b)] and small (but not zero) incident seeding second field [Fig. 4(c)]; in contrast to the generation of two photons, here the propagation equations are not symmetric with respect to the fields. In Fig. 4(b), one can see that initially the intensity of the first field grows quadratically, so the solution describes difference-frequency generation. For the case presented in Fig. 4(c), initially the second field grows exponentially from the seeding. The following hyperbolic growth of the intensities is common for both cases. Again, the solutions in the transition region can be less smooth depending on the initial phases of the fields and phases of the induced susceptibilities.

3. Discussion

Note that processes where q laser photons are split into three photons with *different* frequencies are also possible, as well as HO parametric processes where more than three photons are generated. Such processes can be significant in specific conditions when they are phase matched; they can be described by the corresponding induced susceptibilities, but that study is beyond the scope of this paper.

Finally, we would like to note that to find analytical solutions of the system of Eqs. (24) or the system of Eqs. (26), we assume that the induced susceptibilities do not depend on the propagation distance. Thus we assume that the laser intensity is constant. This assumption can fail due to the laser beam focusing or defocusing, the pulse spreading, as well as due to nonlinear optical effects. On the other hand, the dependence of the induced susceptibility on the laser intensity could be not very pronounced (similarly to the dependence of the HHG efficiency on the laser intensity in certain cases). Thus the study of this dependence is a nice outlook of this research.

4. Stimulated scattering

The schematic of the Stokes wave generation due to the high-order stimulated scattering is presented in Fig. 3. Similar to the perturbative case (see [1], paragraph 10.3), nonlinear polarization can be written as

$$P^{NL}(\omega_1) = \kappa_R^{(1)}(\omega_1)E_1(\omega_1), \quad (27)$$

where $\kappa_R^{(1)}(\omega_1)$ is the induced Raman susceptibility. Note that here we do not specify the number of laser quanta q involved in the process. Practically, $\kappa_R^{(1)}$ can be calculated exactly in the same way as it was done above, namely as the limit of $P(\omega_1,\mathcal{E}_1)/\mathcal{E}_1$ for weak $\mathcal{E}_1$. The propagation equation for the Stokes wave is

$$\left(\frac{\partial}{\partial z} + \alpha\right)\mathcal{E}_1 = i2\pi k_1\kappa_R^{(1)}(\omega_1)\mathcal{E}_1, \quad (28)$$

where α is the linear absorption coefficient for frequency ω_1. Its solution is

$$\mathcal{E}_1 = \mathcal{E}_1(0)\exp\{g_R z - \alpha z\},$$

where $g_R = i2\pi k_1\kappa_R^{(1)}(\omega_1)$. The amplification takes place for $\mathrm{Im}[\kappa_R^{(1)}(\omega_1)] < -\alpha/(2\pi k_1)$. The main practical advantage of this process for effective xuv generation is the absence of the phase-matching problem. Our approach can be useful for describing xuv amplification found in numerical studies [11,41]. Note that the simplified picture presented in Fig. 3 assumes that the excited state is not affected by the laser field, which is a quite questionable approximation. In this connection, it is remarkable that only high-order harmonics near plateau cutoff were amplified in calculations [11,41]. According to the recollision picture [24], these harmonics are emitted at time instant, where the instantaneous laser field strength passes zero, so the approximation of the unaffected excited state can be reliable.

V. CONCLUSIONS

In conclusion, in this paper we suggest the formalism of the nonperturbatively induced susceptibilities, which allows one to write the propagation equations in a form similar (but not identical) to the one used in perturbative nonlinear optics. Under some limitations, we derive the analytical solutions for the propagation equations, describing several high-order optical properties. In particular, the superexponential (hyperbolic) growing solutions are found for the three-photon parametric amplification. The numerically found susceptibilities are too low to make these processes observable in small dilute targets such as gas jets, but in larger targets with higher densities (high-pressure gas-filled capillaries) such processes are feasible, opening a way for the efficient generation of coherent xuv. Finally, we discuss published numerical and experimental results where some processes from the nonperturbative nonlinear optics might have already been observed.

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APPENDIX A: PARAMETRIC GENERATION OF TWO PHOTONS

Let us consider the process in which q photons from the initial field are converted into one ω_1 photon and one ω_2 photon; see Fig. 3(a). In this case, q is an even number (note that for $q = 2$, this is a well-known process of four-wave mixing; however, for higher q, the analogy with the four-wave mixing is hardly helpful). In the plane-wave approximation and slowly varying amplitude approximation, $E_{1,2} = \mathcal{E}_{1,2}(z)\exp\{i(k_{1,2}z - \omega_{1,2}t)\}$ and the complex amplitudes of the fields are described with the following propagation equations:

$$\frac{\partial \mathcal{E}_{1,2}}{\partial z} = \frac{i2\pi\omega_{1,2}^2}{k_{1,2}c^2} P^{NL}(\omega_{1,2},z)\exp\{-i(k_{1,2}z - \omega_{1,2}t)\}, \quad \text{(A1)}$$

where P^{NL} is the nonlinear part of the polarization. For $m = -1$, we have, from Eq. (15),

$$P^{NL}(\omega_{1,2},z) = \kappa_q^{(1)}(\omega_{1,2} = q\omega_0 - \omega_{2,1}) \times \exp\{iq(k_0 z - \omega_0 t + \varphi_0)\}E_{2,1}^*. \quad \text{(A2)}$$

Substituting this equation into Eq. (A1) and assuming in the denominator $k_{1,2} \approx \omega_{1,2}/c$, we have

$$\frac{\partial \mathcal{E}_{1,2}}{\partial z} = i2\pi k_{1,2}\kappa_q^{(1)}(\omega_{1,2})\mathcal{E}_{2,1}^* \exp\{i\Delta k z + iq\varphi_0\}, \quad \text{(A3)}$$

where Δk in the considered case is $\Delta k = qk_0 - k_1 - k_2$.

Propagation equations (A3) are similar to those describing the parametric amplification in the usual (perturbative) nonlinear optics (see [1], part 9.1). Note that this similarity can also be seen from the fact that the perturbative nonlinear response in the presence of the pump field E_3 with the frequency $\omega_3 = \omega_1 + \omega_2$, $P^{NL,\text{per}}(\omega_{1,2}) = \chi^{(2)}E_{2,1}^* E_3$, can be converted in Eq. (A2) substituting $\chi^{(2)}E_3 \to \kappa_q^{(1)}\exp\{iq(k_0 z - \omega_0 t + \varphi_0)\}$. However, an important difference from the perturbative case is that the susceptibility $\kappa_q^{(m)}$ is complex, and its phase is not negligible.

Let us introduce

$$A_{1,2} = i2\pi k_{1,2}\kappa_q^{(1)}(\omega_{1,2}). \quad \text{(A4)}$$

Equations (A3) are rewritten as

$$\frac{\partial \mathcal{E}_{1,2}}{\partial z} = A_{1,2}\mathcal{E}_{2,1}^* \exp\{i\Delta k z + iq\varphi_0\}. \quad \text{(A5)}$$

Below we shall consider the case of the exact phase matching $\Delta k = 0$. Let us introduce

$$\mathcal{E}_{1,2} = u_{1,2}\exp\{i\varphi_{1,2}\}, \qquad A_{1,2} = a_{1,2}\exp\{i\theta_{1,2}\}, \quad \text{(A6)}$$

where $u_{1,2}, \varphi_{1,2}, a_{1,2}, \theta_{1,2}$ are real. Introducing

$$\psi = \theta_1 + q\varphi_0 - \varphi_1 - \varphi_2,$$

$$\Delta\theta = \theta_2 - \theta_1,$$

we have, from Eqs. (A5),

$$\frac{\partial u_1}{\partial z} = a_1 u_2 \cos(\psi), \quad \frac{\partial u_2}{\partial z} = a_2 u_1 \cos(\psi + \Delta\theta), \quad u_1\frac{\partial \varphi_1}{\partial z} = a_1 u_2 \sin(\psi), \quad u_2\frac{\partial \varphi_2}{\partial z} = a_2 u_1 \sin(\psi + \Delta\theta). \quad \text{(A7)}$$

In the absence of resonances, the number of quanta generated at frequencies ω_1 and ω_2 should be equal,

$$\frac{1}{\omega_1}\frac{\partial(u_1^2)}{\partial z} = \frac{1}{\omega_2}\frac{\partial(u_2^2)}{\partial z}.$$

From this equation and the first pair of Eqs. (A7), we find

$$\frac{a_1}{\omega_1}\cos(\psi) = \frac{a_2}{\omega_2}\cos(\psi + \Delta\theta). \quad \text{(A8)}$$

Under $\Delta\theta = 0$, this gives

$$\frac{a_1}{\omega_1} = \frac{a_2}{\omega_2}. \quad \text{(A9)}$$

Under $\Delta\theta \neq 0$, Eq. (A8) can be satisfied only under $\psi =$ const [for every solution of the system of linear equations (A7); for different solutions, ψ can be different]. Thus, $\frac{\partial\psi}{\partial z} = -\frac{\partial\varphi_1}{\partial z} - \frac{\partial\varphi_2}{\partial z} = 0$ and, from the second pair of Eqs. (A7), we find

$$\frac{a_1}{\omega_1} = \frac{a_2}{\omega_2}. \quad \text{(A10)}$$

Thus, this equation is valid irrespectively on $\Delta\theta$. From Eqs. (A4), (A6), and (A10), we have

$$\left|\kappa_q^{(1)}(\omega_1)\right| = \left|\kappa_q^{(1)}(\omega_2)\right|. \quad \text{(A11)}$$

This equation presents the permutation symmetry of induced susceptibilities of the first order. Let us denote $|\kappa_q^{(1)}| = |\kappa_q^{(1)}(\omega_{1,2})|$, so $a_{1,2} = 2\pi k_{1,2}|\kappa_q^{(1)}|$.

The solution of Eq. (A5) is

$$\mathcal{E}_{1,2}(z) = \mathcal{E}^{+}_{1,2}(z) + \mathcal{E}^{-}_{1,2}(z),$$

$$\mathcal{E}^{\pm}_{1}(z) = \mathcal{E}^{\pm}_{1}(0)\exp\{g^{\pm}z + i\varphi^{\pm}_{1}(0)\},$$

$$\mathcal{E}^{\pm}_{2}(z) = \mathcal{E}^{\pm}_{2}(0)\exp\{(g^{\pm})^{*}z + i\varphi^{\pm}_{2}(0)\},$$

$$\mathcal{E}^{\pm}_{1}(0) = \left|\frac{\alpha\mathcal{E}_{2}(0) + \mathcal{E}^{*}_{1}(0)\exp\{i(\theta_1 + q\varphi_0 - \psi^{\pm})\}}{2}\right|,$$

$$\mathcal{E}^{\pm}_{2}(0) = \mathcal{E}^{\pm}_{1}(0)/\alpha,$$

$$\varphi^{\pm}_{2}(0) = \arg\left(\frac{\alpha\mathcal{E}_{2}(0) + \mathcal{E}^{*}_{1}(0)\exp\{i(\theta_1 + q\varphi_0 - \psi^{\pm})\}}{2}\right),$$

$$\varphi^{\pm}_{1}(0) = \theta_1 + q\varphi_0 - \psi^{\pm} - \varphi^{\pm}_{2}(0),$$

where $g^{\pm} = 2\pi\sqrt{k_1 k_2}|\kappa^{(1)}_{q}|\exp i\psi^{\pm}$, $\alpha = \sqrt{\omega_1/\omega_2}$, $\psi^{+} = -\Delta\theta/2$, and $\psi^{-} = -\Delta\theta/2 + \pi$.

For the phase-matched process with $\omega_1 = \omega_0/q$, we have

$$|\mathrm{Re}(g^{\pm})| = 2\pi k_0\left|\kappa^{(1)}_{q}\right||\cos(\Delta\theta/2)|. \tag{A12}$$

APPENDIX B: PARAMETRIC GENERATION OF THREE PHOTONS

The nonlinear polarizations for this process are

$$P^{NL}(\omega_1) = \kappa^{(2)}_{q}(\omega_1 = q\omega_0 - \omega_1 - \omega_2) \times \exp\{iq(k_0 z - \omega_0 t + \varphi_0)\}E^{*}_{1}E^{*}_{2}, \tag{B1}$$

$$P^{NL}(\omega_2) = \kappa^{(2)}_{q}(\omega_2 = q\omega_0 - 2\omega_1) \times \exp\{iq(k_0 z - \omega_0 t + \varphi_0)\}(E^{*}_{1})^{2}. \tag{B2}$$

Let us denote

$$A_1 = i2\pi k_1\kappa^{(2)}_{q}(\omega_1 = q\omega_0 - \omega_1 - \omega_2), \tag{B3}$$

$$A_2 = i2\pi k_2\kappa^{(2)}_{q}(\omega_2 = q\omega_0 - 2\omega_1). \tag{B4}$$

The propagation equations are written as

$$\frac{\partial\mathcal{E}_1}{\partial z} = A_1\mathcal{E}^{*}_{1}\mathcal{E}^{*}_{2}\exp\{i\Delta kz + iq\varphi_0\}, \tag{B5}$$

$$\frac{\partial\mathcal{E}_2}{\partial z} = A_2(\mathcal{E}^{*}_{1})^{2}\exp\{i\Delta kz + iq\varphi_0\}, \tag{B6}$$

where $\Delta k = qk_0 - 2k_1 - k_2$. Below we shall consider the case of the exact phase-matching $\Delta k = 0$.

Let us introduce

$$\mathcal{E}_{1,2} = u_{1,2}\exp\{i\varphi_{1,2}\},$$
$$A_{1,2} = a_{1,2}\exp\{i\theta_{1,2}\}, \tag{B7}$$

where $u_{1,2}, \varphi_{1,2}, a_{1,2}, \theta_{1,2}$ are real. Introducing

$$\psi = \theta_1 + q\varphi_0 - 2\varphi_1 - \varphi_2,$$

$$\Delta\theta = \theta_2 - \theta_1,$$

we have, from Eqs. (B5) and (B6),

$$\frac{\partial u_1}{\partial z} = a_1 u_1 u_2\cos(\psi),$$
$$\frac{\partial u_2}{\partial z} = a_2 u^{2}_{1}\cos(\psi + \Delta\theta),$$
$$u_1\frac{\partial\varphi_1}{\partial z} = a_1 u_1 u_2\sin(\psi),$$
$$u_2\frac{\partial\varphi_2}{\partial z} = a_2 u^{2}_{1}\sin(\psi + \Delta\theta). \tag{B8}$$

In the absence of resonances, the number of quanta generated at the frequency ω_1 should be twice that of quanta at the frequency ω_2,

$$\frac{1}{\omega_1}\frac{\partial\left(u^{2}_{1}\right)}{\partial z} = \frac{2}{\omega_2}\frac{\partial\left(u^{2}_{2}\right)}{\partial z}.$$

From this equation and the first pair of Eqs. (B8), we find

$$\frac{a_1}{\omega_1}\cos(\psi) = 2\frac{a_2}{\omega_2}\cos(\psi + \Delta\theta). \tag{B9}$$

Case 1: $\Delta\theta \neq 0$. Equation (B9) can be satisfied only under $\psi = \mathrm{const}$:

$$\tan(\psi) = \left[\cos(\Delta\theta) - \frac{\left|\kappa^{(2)}_{q}(\omega_1)\right|}{2\left|\kappa^{(2)}_{q}(\omega_2)\right|}\right]\frac{1}{\sin(\Delta\theta)}. \tag{B10}$$

Thus,

$$\frac{\partial\psi}{\partial z} = -2\frac{\partial\varphi_1}{\partial z} - \frac{\partial\varphi_2}{\partial z} = 0, \tag{B11}$$

and from this equation and the second pair of Eqs. (B8), we find

$$u_1(z)/u_2(z) = \mathrm{const}. \tag{B12}$$

The solution of the first pair of Eqs. (B8) satisfying (B12) is

$$u_{1,2}(z) = u_{1,2}(0)\frac{1}{1 - z/z_c}, \tag{B13}$$

where

$$z_c = \frac{1}{u_2(0)a_1\cos(\psi)} \tag{B14}$$

and

$$\frac{u_1(z)}{u_2(z)} = \sqrt{\frac{a_1\cos(\psi)}{a_2\cos(\psi + \Delta\theta)}}.$$

Taking into account equation (B9), this equation can be rewritten as

$$\frac{u_1(z)}{u_2(z)} = \sqrt{\frac{2\omega_1}{\omega_2}}. \tag{B15}$$

Thus, only if the initial conditions satisfy this equation, the solution satisfies Eq. (B12). Finally, from Eqs. (B11), (B15), and the second pair of Eqs. (B8), we find

$$\cos(\Delta\theta) = \frac{\left|\kappa^{(2)}_{q}(\omega_1)\right|}{\left|\kappa^{(2)}_{q}(\omega_2)\right|} - \frac{2\left|\kappa^{(2)}_{q}(\omega_2)\right|}{\left|\kappa^{(2)}_{q}(\omega_1)\right|}. \tag{B16}$$

Thus we find the connection between phases and absolute values of the second-order induced susceptibilities.

Case 2: $\Delta\theta = 0$. Equation (B9) is satisfied for every ψ; it is satisfied under

$$\frac{a_1}{\omega_1} = 2\frac{a_2}{\omega_2}. \tag{B17}$$

So,

$$\left|\kappa_q^{(2)}(\omega_1)\right| = 2\left|\kappa_q^{(2)}(\omega_2)\right|. \tag{B18}$$

This equation presents the permutation symmetry of the induced susceptibilities of the second order for $\Delta\theta = 0$. Note that Eq. (B16) turns to (B18) when $\Delta\theta = 0$.

Let us denote

$$\mathcal{E}_{1,2}(z) = \mathcal{E}'_{1,2}(z)\exp\{i\varphi_{1,2}(0)\},$$

so that $\mathcal{E}'_{1,2}(0)$ are real. Then, Eqs. (B5) and (B6) can be written as

$$\frac{\partial \mathcal{E}'_1}{\partial z} = a_1 \mathcal{E}_1'^{*}\mathcal{E}_2'^{*}\exp\{i\psi(0)\}, \tag{B19}$$

$$\frac{\partial \mathcal{E}'_2}{\partial z} = a_2 (\mathcal{E}_1'^{*})^2\exp\{i\psi(0)\}, \tag{B20}$$

where $\psi(0) = \theta_1 + q\varphi_0 - 2\varphi_1(0) - \varphi_2(0)$ (note that $\theta_1 = \theta_2$ because we consider the case $\Delta\theta = 0$). If initial phases $\varphi_1(0)$ and $\varphi_2(0)$ are chosen so that $\psi(0) = 0$, the solution of Eqs. (B19) and (B20) remains real. Substituting

$$\mathcal{E}'_1(z) = \sqrt{\frac{2\omega_1}{\omega_2}[\mathcal{E}'_2(z)^2 - \widetilde{\mathcal{E}}^2]}, \tag{B21}$$

where $\widetilde{\mathcal{E}} = \sqrt{\mathcal{E}'_2(0)^2 - \frac{\omega_2}{2\omega_1}\mathcal{E}'_1(0)^2}$, the system of equations (B19) and (B20) is presented as a single equation [note also Eq. (B17)]:

$$\frac{\partial \mathcal{E}'_2}{\partial z} = a_1\left(\mathcal{E}_2'^2 - \widetilde{\mathcal{E}}^2\right). \tag{B22}$$

Such equation is known as the Riccati equation. Its solution is

$$\mathcal{E}'_2(z) = \widetilde{\mathcal{E}}\ \frac{\mu\exp\{-\tilde{g}z\} + \exp\{\tilde{g}z\}}{\mu\exp\{-\tilde{g}z\} - \exp\{\tilde{g}z\}}, \tag{B23}$$

where $\tilde{g} = a_1\widetilde{\mathcal{E}}$,

$$\mu = \frac{\mathcal{E}'_2(0) + \widetilde{\mathcal{E}}}{\mathcal{E}'_2(0) - \widetilde{\mathcal{E}}}.$$

Thus, Eqs. (B21) and (B23) give the solution. The typical feature of this solution is a hyperbolic growth after a certain propagation distance,

$$\mathcal{E}'_{1,2}(z) \propto \frac{1}{1 - z/z_c}.$$

The solution goes to infinity at the "catastrophe" distance

$$z_c = \frac{\ln(\mu)}{2\tilde{g}}. \tag{B24}$$

Thus, we can see that in both cases $\Delta\theta \neq 0$ and $\Delta\theta = 0$, there is a hyperbolic growth of the solution after a certain propagation distance.

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