Periodic density modulation for quasi-phase-matching of optical frequency conversion is inefficient under shallow focusing and constant ambient pressure

Itai Hadas and Alon Bahabad*

Department of Physical Electronics, School of Electrical Engineering, Iby and Aladar Fleischman Faculty of Engineering, Tel-Aviv University, Tel-Aviv 69978, Israel
*Corresponding author:alonb@eng.tau.ac.il

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The two main mechanisms of a periodic density modulation relevant to nonlinear optical conversion in a gas medium are spatial modulations of the index of refraction and of the number of emitters. For a one-dimensional model neglecting focusing and using a constant ambient pressure, it is shown theoretically and demonstrated numerically that the effects of these two mechanisms during frequency conversion cancel each other exactly. Under the considered conditions, this makes density modulation inefficient for quasi-phase-matching an optical frequency conversion process. This result is particularly relevant for high-order harmonic generation.

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Generally, optical frequency conversion processes suffer from low conversion efficiency due to a dispersion-induced-phase mismatch between the generated signal and the pump laser. Quasi-phase-matching (QPM) [1] is a well-known technique that applies a macroscopic modulation over a relevant parameter of the nonlinear interaction. This systematically corrects the phase mismatch and hence significantly increases the frequency conversion efficiency. Nonlinear optics in gas phase is particularly relevant to the extreme nonlinear process of high-order harmonic generation (HHG) [2–4], which became a valuable research resource in various areas such as spectroscopy, material characterization, and microscopy.

In the last two decades various experiments have successfully demonstrated QPM for HHG. Rightfully, they applied the modulation to both the amplitude (through the number of emitters) of the nonlinear polarization and to its phase through the index of refraction. Approximations (or rather neglecting some terms) during the analysis led to the conclusion that QPM is possible using density modulation in a focused beam geometry. In 2007 Auguste et al. [12] considered numerically sinusoidal density modulation accompanied by both a focused pump beam and a displaced truncated Lorentzian ambient pressure profile. Their model suggested intensity buildup due to density modulation. In 2007 Seres et al. [13] conducted an experiment in which two consecutive and separated gas jets were used as the medium for HHG and showed in-phase emission. We note that having only two sources does not amount to a periodic modulation per se. Furthermore, various mechanisms in this case can bring the emission from the two sources into phase matching. Examples are axial intensity differences (which influence the phase of the emitted harmonics [14]) and Gouy phase shift, both due to diffraction; these mechanisms are independent from the density modulation associated with the existence of the two sources.

In 2012 Sapaev et al. [15] suggested an ultrasound pressure modulation to quasi-phase-match third harmonic generation (THG) in gas phase. However, in that work modulation of the index of refraction was only partially accounted for, while its effect on the phase mismatch term was neglected.

In this Letter we prove analytically for a one-dimensional model that, when no terms are neglected in the analysis, a sinusoidal density modulation over a constant ambient pressure leads to amplitude and phase modulations of the nonlinear polarization that cancel each other for the supposed phase-match component. Simulations suggest our results are relevant to more general (not only sinusoidal) periodic density modulations. These results are relevant for shallow focusing (in particular when the Rayleigh length of the pump beam is much longer than the coherence length of the interaction) or for waveguide geometry.

Our analytical model is one-dimensional for an upconversion process using an undepleted pump the evolution of the generated harmonic amplitude is given by [16]
Here \( g \) designates the harmonic order being generated. The prefactor is given with 
\[ K = -in_0^p [e^{iK(z)/2}] \times d_\text{eff} \times E_0(z), \]
where \( n_0 \) is the angular frequency of the generated radiation, \( k \) is the wave-vector, \( d_\text{eff} \) is a nonlinear coupling coefficient, \( E_0 \) is the pump field amplitude, and \( p \) is a positive integer. We assume the pump field has constant amplitude, which is justified for an interaction length shorter than the Rayleigh range of the pump beam under non-depletion approximation. The wave-vector itself, \( k(n_0) \), is also modulated by the density modulation. As this is typically a weak modulation (especially for HHG) we can consider \( k^{-1}(n_0) \) appearing in the prefactor \( K \) to be constant (we verified this point with simulations). Overall, we consider \( K \) to be constant in \( z \). The phase of the nonlinear polarization is given with \( \phi(z) = \int_0^z \Delta k(z')dz' \) where \( \Delta k(z) \) is the phase mismatch of the interaction.

The nonlinear polarization relative amplitude is given with \( g(z) = g_0(1 - m \cos(Kz)) \) where \( g_0 \) is proportional to the unperturbed gas density, \( K \) is the modulation frequency, and \( 0 < m \leq 1 \) is the modulation depth. The density modulation creates a weak modulation in the index of refraction in the form of \( n = 1 + (\alpha(z))P \), where \( \alpha(z) \) contains neutral gas, plasma, and (usually negligible) nonlinear dispersion terms [17]. \( P \) is the gas pressure. Typically \( \alpha(z)P \ll 1 \). Important, however, is to notice that the phase mismatch of the process \( \Delta k \) depends on the \( P \)-dependence of the index at the fundamental and harmonic frequencies, and so it would be modulated at the same modulation depth as the gas density itself: \( \Delta k = \Delta k_0(1 - m \cos(Kz)) \) where \( \Delta k_0 \) is the unperturbed phase mismatch.

Using the above forms for \( g(z) \) and \( \Delta k(z) \) in the evolution, Eq. (1) yields:

\[
\frac{dE}{dz} = K_0 \left(1 - m \cos(\Delta k_0z/d)\right) e^{-i\Delta k_0z/d} e^{i\Delta k_0z/d} 
\]

where we used \( K_0 \equiv \Delta k_0/d \) with \( d \) being the order of the modulation relative to the unperturbed phase mismatch. Achieving QPM requires using a (positive, non-zero) integer valued \( d \) and looking for a term on the right-hand side (RHS) of Eq. (2) that does not oscillate. To do so we first expand the term \( e^{i\Delta k_0z/d} \):

\[
e^{i\Delta k_0z/d} = \sum_{n=0}^{\infty} \frac{(imd/2)^n}{n!} \left( e^{i\Delta k_0z/d} - e^{-i\Delta k_0z/d} \right)^n 
\]

\[
= \sum_{n=0}^{\infty} \sum_{p=0}^{n} \frac{(imd/2)^n}{(n-p)! (p)!} (-1)^p e^{i(n-2p)\Delta k_0z/d} 
\]

To get an exponential term that depends on a single summation index we define \( l \equiv n - 2p \), such that:

\[
e^{i\Delta k_0z/d} = \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} \frac{(imd/2)^n}{(n-l)! (n+l)!} (-1)^l e^{i(n-2p)\Delta k_0z/d} 
\]

\[
= \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} \frac{(imd/2)^n}{(n-l)! (n+l)!} (-1)^l e^{i\Delta k_0z/d} 
\]

(3)

(4)

Here \( (+2) \) indicates that the index \( l \) changes in steps of 2. We can now switch the order of summation using the fact that for each \( l = -\infty, \ldots, \infty \) the range of \( n \) is \( n \geq |l| \) while \( n \) has the same parity as \( l \):

\[
e^{imd \sin(\Delta k_0z/d)} = \sum_{l=-\infty}^{\infty} (-i)^l e^{i\Delta k_0z/d} \sum_{n=0}^{\infty} \frac{(imd/2)^n}{(n-l)! (n+l)!} (-1)^l 
\]

(5)

Using this result back in the evolution equation leads to:

\[
\frac{dE}{dz} = K_0 \left(1 - m \cos(\Delta k_0z/d) - m \cos(\Delta k_0z/d) - \sum_{l=-\infty}^{\infty} (-i)^l e^{i\Delta k_0z/d} \sum_{n=0}^{\infty} \frac{(imd/2)^n}{(n-l)! (n+l)!} (-1)^l \right) 
\]

\[
\times \sum_{l=-\infty}^{\infty} (-i)^l e^{i\Delta k_0z/d} e^{i\Delta k_0z/d} \sum_{n=0}^{\infty} \frac{(imd/2)^n}{(n-l)! (n+l)!} (-1)^l 
\]

(6)

The RHS of this equation can be cast as a sum of three terms \( S_0 + S_1 + S_2 \) where

\[
S_0 = K_0 \sum_{l=-\infty}^{\infty} (-i)^l e^{i\Delta k_0z/d} \sum_{n=0}^{\infty} \frac{(imd/2)^n}{(n-l)! (n+l)!} (-1)^l 
\]

\[
S_1 = -mK_0 \sum_{l=-\infty}^{\infty} (-i)^l e^{i\Delta k_0z/d} \sum_{n=0}^{\infty} \frac{(imd/2)^n}{(n-l)! (n+l)!} (-1)^l 
\]

\[
S_2 = -mK_0 \sum_{l=-\infty}^{\infty} (-i)^l e^{i\Delta k_0z/d} \sum_{n=0}^{\infty} \frac{(imd/2)^n}{(n-l)! (n+l)!} (-1)^l 
\]

(7)

(8)

(9)

To achieve QPM we need a non-zero coefficient multiplying an exponential with a zero frequency. For \( d \)-order QPM we get such an exponential by setting \( l = d \) in \( S_0 \), \( l = d - 1 \) in \( S_1 \), and \( l = d + 1 \) in \( S_2 \). The coefficient for this zero-frequency exponential is given by gathering the appropriate terms from \( S_0, S_1, \) and \( S_2 \):

\[
I = K_0 \left[ (-i)^d \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} \frac{(imd/2)^n}{(n-l)! (n+l)!} \frac{m}{2} (-i)^{d-1} \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} \frac{(imd/2)^n}{(n-l)! (n+l)!} 
\]

\[
\times \frac{m}{2} (-i)^d + \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} \frac{(imd/2)^n}{(n-l)! (n+l)!} \frac{m}{2} (-i)^{d+1} \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} \frac{(imd/2)^n}{(n-l)! (n+l)!} \right] 
\]

(10)

To proceed we substitute \( n \to n - 1 \) for the second sum and \( n \to n + 1 \) for the third sum:

\[
I = K_0 \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} \frac{(-i)^d (imd/2)^n}{(n-l)! (n+l)!} 
\]

\[
- (-i)^d \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} \frac{m}{2} (imd/2)^{n+1} \frac{m}{2} (-i)^{d+1} \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} \frac{m}{2} (imd/2)^{n+1} \frac{m}{2} (-i)^{d+1} \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} \frac{m}{2} (imd/2)^{n+1} \frac{m}{2} (-i)^{d+1} \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} \frac{m}{2} (imd/2)^{n+1} 
\]

(11)

The first two sums start with terms proportional to \( m^d \) while the third sum starts with a term proportional to \( m^{d+2} \). As such, we set \( I = I_1 + I_2 \) where \( I_1 \) will sum the terms proportional to \( m^d \), and \( I_2 \) will sum all other terms:

\[
I_1 = K_0 \left[ (-i)^d \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} \frac{m}{2} (imd/2)^{n+1} \frac{m}{2} (-i)^{d+1} \sum_{l=0}^{\infty} \sum_{n=0}^{\infty} \frac{m}{2} (imd/2)^{n+1} \right] = 0 
\]

(12)
The coefficient of the (supposedly) phase-matching term is $I = 0$ for any QPM order, $d$, and for any pressure modulation depth, $m$. Thus QPM cannot be achieved using a sinusoidal density modulation. We note that the exact cancellation occurred because we had both amplitude and phase modulation of the nonlinear polarization with the same modulation depth.

To corroborate our analytical result we have performed two numerical simulations. The first, simple one, integrates Eq. (1) using the Runge-Kutta 4 (RK4) method. The second simulation solves a one-dimensional model of HHG with the following parameters: the pump is a 25 fs pulse at a center wavelength of 800 nm with peak intensity of $1.5 \times 10^{14}$ W/cm$^2$ corresponding to low ionization of 1.44%. The gas medium is made of Argon atoms at an ambient pressure of $P_0 = 300$ Torr at 297 K corresponding to atomic ambient density of $9.75 \times 10^{18}$ cm$^{-3}$. The single atom interaction is simulated by calculating the polarization acceleration through numerical integration of the time-dependent Schrödinger equation (TDSE) using the symmetric split-step Fourier method [18]. Propagation of the pump beam is calculated by RK4 integration of a propagation equation given by Geissler et al. [19] to which a neutral atom dispersion term was added. The overall

$$I_2 = k g_0 \sum_{n=0}^{\infty} \left[ (-i)^d \frac{\text{imd}(/2)^n}{(\frac{n+d}{2})!} - (-i)^{d+1} \frac{\text{imd}(/2)^{n+1}}{2 (\frac{n+d+2}{2})!} \right]$$

We now substitute $n \rightarrow n + 2$ in the first sum to get:

$$I_2 = (-i)^d k g_0 \sum_{n=0}^{\infty} \left[ \frac{(\text{imd}(/2)^{n+2}}{(\frac{n+d+2}{2})!} - \frac{im (\text{imd}(/2)^{n+1}}{2 (\frac{n+d}{2})! (\frac{n+d+2}{2})!} \right]$$

$$+ \frac{im (\text{imd}(/2)^{n+1}}{2 (\frac{n+d}{2})! (\frac{n+d+2}{2})!} = (-i)^d k g_0 \sum_{n=0}^{\infty} \frac{\text{imd}(/2)^{n+2}}{(\frac{n+d+2}{2})! (\frac{n+d+2}{2})!}$$

$$\times \left[ d - \frac{n + d + 2}{2} + \frac{n - d + 2}{2} \right] = 0. \quad (13)$$

Fig. 1. Simulations for optical frequency conversion with density modulation. Columns: (a) sinusoidal pressure modulation with 10% modulation depth for 300 Torr ambient pressure; (b) Gaussian train pressure modulation at a 100% modulation depth for 300 Torr peak pressure; and (c) Super-Gaussian train pressure modulation at a 100% modulation depth for 300 Torr peak pressure. Rows: (I) form of the modulation function; (II) evolution of the intensity of the up-converted field from the simple model—squared absolute value of the integration result of Eq. (1); and (III) evolution of the envelope of the 25th harmonic from the HHG numerical model. For the evolution graphs: continuous (blue) line—non modulated medium; dashed (red) line—amplitude-only (number of emitters) modulation of the nonlinear polarization; dashed–dotted (magenta) line—phase-only (index of refraction) modulation of the nonlinear polarization; and dotted (black) line—full density modulation acting on both the number of emitters and on the index of refraction.
numerical model contains a pressure-dependent index of refraction, with contributions from both the free-electrons (plasma) and the neutral atoms in the medium.

The above pump peak power and atomic density corresponds to coherence length of \( l_c = \pi/\Delta k_0 = 174 \mu m \) for the generation of the 25th harmonic. The overall interaction length in the simulations was 3 mm. The value for the unperturbed generation of the 25th harmonic. The overall interaction length was used in both simulations.

The simulation output is the evolution of the 25th harmonic radiation along the propagation axis. We considered three types of periodic modulations: a sinusoidal, a sequence of Super-Gaussians (with an exponential power of \( \alpha c^2 \) and width of \( l_c/2 \)). The sinusoidal and Gaussian modulations had a period of \( 2l_c \) (QPM order \( d = 1 \)), and the super-Gaussian modulation had a period of \( 4l_c \) (QPM order \( d = 2 \)). The sinusoidal modulation depth was \( m = 0.1 \) while the other two modulations were at 100% modulation depth.

Simulation results are shown in Fig. 1. For the simple model an amplitude modulation of the nonlinear polarization leads to efficient QPM, while phase modulation of the nonlinear polarization can lead to significant buildup of the harmonic radiation depending on the length of the interaction and exact modulation. For the HHG simulations, phase modulation seems more efficient for modulations which represent isolated gas sources, while amplitude modulation is more efficient for the sinusoidal case. However, in both models, a realistic density modulation that leads to both amplitude and phase modulations together yields no better results than no modulation at all, in accord with the analytical model.

To conclude, application of a periodic density modulation has been previously proposed for quasi-phase-matching optical frequency conversion in a gas medium [11–13,15]. In this Letter, for a one-dimensional model neglecting focusing, pump depletion and using a constant ambient pressure, it was analytically proven that a sinusoidal density modulation cannot lead to QPM as the effects of modulation in the indices of refraction and in the density of radiating emitters would cancel each other. Further simulations numerically integrating our simple model equation as well as a more complete realistic (including pump propagation effects such as depletion, yet still one-dimensional neglecting focusing) model of HHG corroborate our findings for different periodic density modulations. This proves the inefficiency of QPM in gas phase using density (pressure) modulation in a waveguide or under shallow focusing conditions.

This conclusion, however, does not eliminate the possibility of applying QPM using density modulation when further effects are accounted for. An example is interplay between focusing and different gas profiles [12] or the inclusion of strong Kerr nonlinearity leading to additional intensity modulation, which in turn modulates the phase of the emitted harmonics during HHG [14].

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