Quasi-phase-matching and dispersion characterization of harmonic generation in the perturbative regime using counterpropagating beams

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Abstract: It is shown theoretically that second harmonic generation can be quasi-phase-matched by using a pump beam consisting of a forward propagating field and a counterpropagating pulse train. The counterpropagating setup can also be used for direct measurement of the coherence length of the nonlinear process which can determine the dispersion properties of the medium.

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OCIS codes: (190.0190) Nonlinear optics; (190.2620) Harmonic generation and mixing.

References and links

1. Introduction

The efficiency of nonlinear optical processes involving beams with different frequencies is often limited because of dispersion [1]. Different frequency components in the nonlinear process propagate with different phase velocities, restricting efficient energy transfer to a coherence length ($l_c$), which is the distance over which the accumulated phase difference between the signal field and the nonlinear polarization is equal to $\pi$. In quasi phase matching [1, 2], efficient energy transfer is achieved over an extended length using an ordered spatial modulation of one of the interaction parameters, making it possible to sequentially correct for the phase mismatch. For low-energy, low-order (second, third, etc.) harmonic generation processes, the parameter that is modulated is typically the linear [3, 4] or (more commonly) the nonlinear electric susceptibility. This modulation can be implemented using either fabrication pre-processing (e.g. by electric field poling of ferroelectric crystals [5]) or by optical induction in a photorefractive medium [6]. Several approaches for implementing quasi-phase matching based on grating structures have been implemented to date, which also allow for tailoring of the spectral and spatial properties of the nonlinear response [2, 7]. It was also suggested that phase matching can be achieved through the periodic intensity modulation which occurs when light propagates within a specifically-tailored waveguide directional coupler [8].

In 1995 Peatross et al. [9] suggested that weak counterpropagating light beams can be used to manipulate the “extreme” nonlinear optical process of high harmonic generation (HHG). In high harmonic generation, the nonlinear medium is ionized by an intense light pulse. Free electrons ionized from individual atoms or molecules are accelerated by the oscillating laser field and some fraction of these electrons can recombine with their parent ion, releasing their excess kinetic energy in the form of high energy photons [10]. During the trajectories of these unbounded electrons, they are extremely sensitive to changes in the accelerating field. It was predicted and subsequently demonstrated that even a relatively weak counterpropagating pulse - i.e. orders of magnitude less intense than the driving laser and too weak to influence ionization of the medium - suppresses harmonic emission when interfering with the forward driving pulse [11, 12]. Essentially the standing wave oscillations of the pump beam are strongly amplified during the generation of high harmonics, causing a severe phase mismatch that suppresses the high harmonic yield. The influence of the relative polarization between the driving laser and the counterpropagating beam on HHG suppression was also explored [13]. Taking advantage of this suppression effect, quasi phase matching of HHG can be induced...
optically by using a sequence of counterpropagating pulses interfering with the forward propagating beam in out-of-phase coherent zones [12, 14]. This technique was recently demonstrated for high harmonic generation in a hollow waveguide [15-17]. Counterpropagating pulses were also used for local in-situ probing of the coherence length in HHG [18, 19] by continuously changing the place where the counterpropagating beam interferes with the forward propagating beam. Finally, it was also suggested that a weak counterpropagating quasi-CW beam could be used to implement grating-assisted phase matching of HHG [20].

Here we show that all-optical quasi phase matching can also be applied to low-order nonlinear optics, albeit with a different physical picture in this perturbative nonlinear process. As in extreme nonlinear optics, the counterpropagating pulses are used for manipulating the harmonic emission in out-of-phase zones. However, instead of using weak counterpropagating light to change the trajectories of photo-ionized electrons during HHG, the nonlinear polarization induced by bound electrons is modified by a counterpropagating beam with intensity comparable to that of the forward propagating beam. This is to our knowledge a new concept for perturbative nonlinear optics: in addition to the known methods of modulating the linear or nonlinear susceptibility and of periodic light coupling for achieving quasi phase matching of low-order nonlinear processes, optical interference can be used to spatially modulate the nonlinear polarization wave. The merits of such a method include the possibility for complete dynamical real-time wavelength tunability over the entire transparency range of any nonlinear material, with no need for material pre-processing. Moreover, the same scheme can be used to measure in-situ the coherence length with which the linear index of refraction of the material can be extracted at different wavelengths – revealing its dispersion properties. Traditional methods for measuring the index of refraction such as prismatic dispersion [21] or by using an optical three-wave-mixing processes [22] probe homogenous samples and cannot track changes in the index of refraction within the material. In addition, use of femtosecond pulses in white-light interferometry can usually accurately extract only higher order dispersion terms - assuming a homogenous sample [23].

2. Nonlinear polarization and field build-up in the presence of counterpropagating light

To understand how a train of counterpropagating pulses can be used for quasi phase matching, we first need to establish how the nonlinear polarization and signal buildup are influenced by counterpropagating light. Although this approach can be applied to any low-order harmonic generation process, for simplicity we consider here the most basic one – second harmonic generation. The extension to other nonlinear processes is in-general straightforward.

Consider a pump beam consisting of forward and backward propagating fields:

\[
E(z,t) = E_f(z,t)e^{i(\omega t - kz)} + E_b(z,t)e^{i(\omega t + kz)},
\]

where \(E_f(z,t)\) and \(E_b(z,t)\) are the forward and backward field envelopes. We can write the field equivalently in the form of a forward propagating field:

\[
E(z,t) = \left[ E_f(z,t) + E_b(z,t)e^{2iz} \right] e^{i(\omega t - kz)},
\]

which allows the forward-propagating second-order nonlinear polarization wave to be expressed as:

\[
P_{NL}^{(2)}(z,t) = \kappa' \left[ E_f(z,t) + E_b(z,t)e^{2iz} \right] e^{2i(\omega t - kz)},
\]

where \(\kappa' = \varepsilon_0 \chi^{(2)}\) with \(\varepsilon_0\) being the vacuum permittivity and \(\chi^{(2)}\) the relevant component of the second-order nonlinear susceptibility tensor. We now consider an undepleted forward-propagating CW field giving rise to a forward propagating CW second harmonic field. We now assume the backward propagating field is undepleted and has a constant envelope of
form: \( E_b(z,t) = E_{bg}(z + v_g t) \) where \( v_g \) is the group velocity. In this case we can formulate a propagation equation for the envelope of a specific wave front of the second harmonic field:

\[
\left( \frac{\partial^2}{\partial z^2} - 2 i k_{2\omega} \frac{\partial}{\partial z} \right) E_{2w}(z) = \kappa' \left[ E_f + \tilde{E}_b(z)e^{2i\Delta z} \right]^2 e^{i\Delta z} ,
\]

(4)

where \( \Delta k = k_{2\omega} - 2 k_{\omega} \) is the phase mismatch involving the wave vectors of the second harmonic and the pump beam and \( \kappa'' = -8(\omega/c)^2 \chi^{(2)} \) where \( c \) is the speed of light. For a given backward propagating pulse shape with a spatial extension (e.g. FWHM) of length \( L \), the forward propagating wave front would encounter it over a propagation distance of \( L \cdot v_p / (v_p + v_g) = L/2 \), where \( v_p \) is the phase velocity of the forward propagating beam.

In this case, \( \tilde{E}_b(z) = E_b(z \cdot (v_p + v_g) / v_p, t = t_0) \) is the envelope encountered by a specific phase front of the forward propagating field. The value of \( t_0 \) would be different for different phase fronts.

To gain more insight and to be able to derive some analytical results we would like to adopt the slowly-varying envelope approximation (SVEA) i.e. neglecting the second derivative in Eq. (4). Note that in the places where the forward and backward propagating fields intersect, the total pump amplitude corresponds to a standing wave with a period of \( \lambda/2 \). One might expect that these relatively fast spatial oscillations can violate the condition needed for adopting the SVEA. However, as we shall see below, provided that the backward propagating pulses are smooth and we are only interested in the forward propagating second-harmonic field, inaccuracies introduced by using the SVEA will not be large. When the SVEA is adopted, Eq. (4) can be integrated over an interaction length \( L \) to yield:

\[
E_{2w}(z = L) = \kappa \int_{z=0}^{L} \left[ E_f + \tilde{E}_b(z)e^{2i\Delta z} \right]^2 e^{i\Delta z} dz ,
\]

(5)

where \( \kappa = -4 i(\omega/c)^2 \chi^{(2)}/k_{2\omega} \).

If we now consider the interactions between forward and backward CW waves \( E_f(z,t) = E_f \) and \( E_b(z,t) = E_b \) over an interaction length of half a wavelength we get:

\[
E_{2w}(z = \lambda/2) = \kappa E_f^2 \sum_{m=0}^{2} \left( \frac{2}{m} \right)(E_b / E_f)^m \int_{z=0}^{\pi/k} e^{i\Delta z} e^{2i\Delta z} dz .
\]

(6)

If there is no phase mismatch (\( \Delta k = 0 \)), the only non-vanishing term is given for \( m = 0 \).

The integration of Eq. (6) in this case yields the same value as if we started with no backward propagating field in Eq. (5) (which is now proportional to the integration of the nonlinear polarization wave). This result means that under phase-matching condition, the influence of the counterpropagating light on the second harmonic signal at the output of a nonlinear medium with \( L >> \lambda \) is negligible. This result is true for all other perturbative nonlinear polarization terms \( P_{NL}^{(p)}(z,t), \) where \( p \) is a small integer. We note that this situation is different than in the case explored by Peatross et. al. for high harmonic generation, where the nonlinear polarization term scales with different powers for the amplitude and phase of the fundamental field [24].

When the process is not phase matched (\( \Delta k \neq 0 \)), radically different behavior arises. If we assume for simplicity that the coherence length is some multiple of half the fundamental
wavelength \( l_c = \pi / \Delta k = N \cdot \lambda / 2 \) (a realistic scenario when we allow for wavelength and temperature tuning of a specific SHG process), the required ratio between the backward and forward propagating field amplitude \( E_b / E_f \) can be solved exactly to achieve either a total of zero nonlinear response or enhancement of the SHG signal, in those zones where the SHG signal would be otherwise suppressed when there is only a forward propagating beam. To find the \( E_b / E_f \) value for a total of zero nonlinear response Eq. (6) is equated to zero. To find the \( E_b / E_f \) value for an enhancement of the SHG signal Eq. (6) is equated to minus the value given by Eq. (5) when \( E_b = 0 \). For both cases we only need to find the zeros of a quadratic equation. This equation depends on the ratio between the coherence length and the fundamental wavelength and it would always have a solution if \( E_b / E_f \) is allowed to be complex. A similar analysis for another perturbative nonlinear polarization term \( P_{NL}^{(p)}(z,t) \) would amount to finding the zeros of a p-order polynomial equation.

To find the time dependence of the second harmonic field at the end of the interaction length we note that different phase fronts would encounter translated counterpropagating fields. Considering the same interaction length, if for a given wave front the encountered counterpropagating envelope is \( \tilde{E}_b(z) \) then, using Eq. (1) and Eq. (2), for a wavefront delayed by \( z_0 = v_p t_0 \) the envelope encountered would be \( \tilde{E}_b(z + z_0 v_p / v_p) e^{i k z_0} \). The forward phase front amplitude, in this case, would be \( E_f e^{i k z} \).

Plugging this into Eq. (5) while changing the parameter \( t_0 \) into the time variable \( t \) gives:

\[
E_{2w}(z = L, t) = N e^{2i \alpha} \int_{z_0}^{L} \left[ E_f + \tilde{E}_b(z + tv_p) e^{2i k z} \right] e^{i \Delta k z} dz ,
\]

where \( \omega = k v_p \).

Opening the square brackets and changing variables \( \tilde{z} = z + tv_g \) we find oscillating terms at the angular frequencies \( 2 \omega, 2 \omega(1 - v_g / v_p) - \Delta k \cdot v_g \) and \( 2 \omega(1 - 2v_g / v_p) - 2 \Delta k \cdot v_g \).

3. Quasi phase matching of low-order nonlinear processes using counterpropagating pulses

The ability to eliminate or enhance the SHG emission over a coherence length using a counterpropagating field enables quasi phase matching using a periodic sequence of counterpropagating pulses (see Fig. 1). We first consider a train of counterpropagating square pulses, such that the interaction of each pulse with the forward propagating field extends over one coherence length. The spacing between the pulses is such that the forward propagating field encounters the pulses every two coherence lengths. The amplitude ratio \( E_b / E_f \) is calculated using the procedure discussed after Eq. (6). Simulations of the growth of the SHG amplitude are shown in Figs. 2(a) and 2(b) for a coherence length \( l_c = 5 \Delta \lambda / 2 \). The bold (red) line represents the second harmonic growth using Eq. (5) (with the SVEA.) The solid (blue) line is the result of a simulation using Eq. (4) (without assuming the SVEA). The dashed line plots the SHG signal without a counterpropagating field.
In the simulations shown in Fig. 2(a), the ratio between the amplitudes of the backward and forward propagating fields is set such that the total harmonic emission in the intersection region is zero. As a result, the SHG amplitude at the end of the section where the backward field is present is the same as at the beginning of the section (see the red line in the inset). This is equivalent to the case when there is only a forward propagating field present, and the nonlinear response is switched on and off with a periodicity of $2l_c$, as for example is the case for periodically-poled fused silica [25]. We can regard this case as an elimination of the nonlinear response in the overlap regions of the relevant phase front with the counterpropagating pulses.

In the simulations shown in Fig. 2(b), the field ratio is set to invert the phase of the nonlinear response in the intersection region. This gives higher efficiency than the previous case. Here the value of the field amplitude at the end of a section where the backward propagating field is different than zero is the same as if there was only a forward propagating field and the nonlinear response of the medium changes polarity every coherence length. This is the case, for example, with quasi-phase-matching using periodically poled ferroelectric crystals [5].

Calculations both assuming and not assuming the SVEA predict that the second harmonic radiation, averaged over several coherence lengths, builds-up continuously along the interaction length when the counterpropagating field is applied. The simulations not assuming the SVEA show large amplitude oscillations that result from the sharp modulations of the counterpropagating pulses. If smoother pulses are used (rather than pulses with a rectangular profile), the differences between the predictions with and without the SVEA diminish significantly. In Figs. 2(c) and 2(d), super-Gaussian pulses of order 20 are used without the SVEA, with amplitude ratio between the backward and forward propagating fields determined by the corresponding analysis for rectangular pulses while using the SVEA. For Fig. 2(c), the coherence length is $l_c=14\lambda/2$ while for Fig. 2(d) $l_c=7\lambda/2$. For both cases, the super Gaussian pulse widths are $0.7l_c$. The increase in the second harmonic build up is much smoother than in the case of rectangular pulses. However, because the specific choice of amplitude ratio is optimized for rectangular pulse profile the efficiency of the process is reduced. The evolution of the second harmonic envelope is sensitive to the detailed parameters of the counterpropagating pulses – the amplitude, width and shape of each pulse.
4. Probing the dispersion of the nonlinear medium

In past work, the use of counterpropagating light pulses for influencing the nonlinear polarization emission in HHG enabled probing the local coherence length and its dynamics within the nonlinear medium [18, 19]. Below, we show that the coherence lengths of low-order processes can be measured by using counterpropagating pulses. Moreover, because the relation between the coherence length and dispersion properties is much simpler for SHG than for HHG, using numerical methods, it is possible to directly extract the refractive index from measurements of the coherence length at different wavelengths and temperatures [22].

The setup we consider here consists of a forward propagating pulse and two counterpropagating pulses, each with a spatial extent less than a coherence length. The delay between the two counterpropagating pulses is varied continuously while the power of the generated forward second harmonic pulse is measured at the exit of the medium. We simulated such a scenario using coupled wave equations (employing the SVEA) for nonlinear interaction of pulses [26]. In this case the equation governing the space and time evolution of the envelope of the second harmonic pulse is:
\[
\left( \frac{\partial}{\partial z} + \frac{1}{v_{g2}} \frac{\partial}{\partial t} \right) E_{2w}(z,t) = \kappa \left[ E_f(z,t) + E_b(z,t) e^{2ikz} \right] e^{i\Delta k z},
\]

where \( v_{g2} \) is the group velocity of the second harmonic pulse. This form assumes that group velocity dispersion can be neglected. Under the assumption that the pump beam is undepleted and that the group velocity of all the pulses is the same, we numerically solved this equation by using a symmetric split step procedure [27]. Because of the use of the SVEA, the pulse shape was chosen to be Super Gaussian of order 40 rather than rectangular. The coherence length was chosen to be \( l_c = 15\lambda/2 \). The pulse width of each pulse was \( l_c/2 \). The interaction length was chosen to be \( 9l_c \). The required ratio between the backward and forward propagating field amplitude for enhancement of the SHG signal (in regions where backward and forward propagating pulses intersect) was estimated using the procedure discussed after Eq. (6).

When the separation between the counterpropagating pulses is equal to multiples of \( 4l_c \) (twice the coherence length in the frame of the forward propagating pulse) a maximal enhancement of the second harmonic amplitude is observed (see Fig. 3(a)). This corresponds to the spacing required for quasi phase matching. When the spacing equals an odd number of coherence lengths in the frame of the forward propagating pulse, the contribution of the two counterpropagating pulses cancel (see Fig. 3(b)). The periodicity of the second harmonic amplitude as a function of the counterpropagating pulses separation is \( 4l_c \) (see Fig. 3(c)). As additional counterpropagating pulses are added, the fidelity of the periodic signal would be improved, allowing for an easier detection of the coherence length. Shorter pulses, higher super Gaussian orders and a smaller ratio between the coherence length and the wavelength would all result in a smoother and higher fidelity curve, although those conditions are harder to produce experimentally.

Note that although our simulations neglect group velocity mismatch (GVM), in reality the difference between the group velocities need only be small enough such that any walk-off between the pump and the generated second-harmonic is less than their pulsewidth after propagation through several coherence lengths. If, however, there is a need to probe the coherence length over extended lengths and GVM is severe, a more elaborate scheme would be needed - for example by locking to the temporal slice of the second harmonic signal which resides under the forward propagating pump envelope.
Fig. 3. Extracting the coherence length of an SHG process using a forward propagating super Gaussian pulse and a pair of two counterpropagating super Gaussian pulses spaced by varying intervals. The super Gaussian order of all pulses is 40. The width of all pulses is $l_c/2$, while $l_c=15\lambda/2$ and $E_b/E_f=9$. The small captions schematically depict the relative spatial pulse separation in the moving frame of the forward propagating pulse. The ± regions are one coherence length long. All (+) regions contribute in-phase to the SHG signal, and similarly for the (–) regions, while the (+) and (–) regions are out of phase with each other. (a) Separation between the counterpropagating pulses is 4$l_c$. (b) Separation is 6$l_c$. (c) Varying the separation continuously results in a periodic signal of period 4$l_c$. (Note a.u. signifies arbitrary units).

5. Summary and conclusions

We have shown theoretically that quasi phase matching of second harmonic generation can be achieved by periodically modulating the pump field with a counterpropagating field. In principle this method can be extended to higher-order nonlinear processes (third harmonic generation etc.). This scheme provides an alternative to the traditional modulation of the nonlinear medium with potential advantages. This approach can be tuned in wavelength over all of the transparency range of the medium, and does not require fabrication of the nonlinear medium. Dynamical changes in the pulse train and complex spatial ordering might lead to interesting applications such as multiple harmonic generation, all-optical control and pulse shaping of the generated harmonics. Finally, we have shown that it is also possible to probe the index of refraction of the nonlinear medium over a very wide spectral bandwidth using counterpropagating pulses, without subjecting the medium to any significant preprocessing.

Acknowledgments

We gratefully acknowledge support for this work from the NSF Center for Extreme Ultraviolet Science and Technology.