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Enhancement of third harmonic generation by wave vector mismatch to counter phase-modulation

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Abstract

Recent experimental developments in material sciences have generated hope that it will be possible to devise optical media where the difference in group velocity between the fundamental and third harmonic may be strongly suppressed. Under these circumstances both pulses would travel together over a long distance. This would lead to an enhancement of the generation process, and hence strong focusing and/or using ultra-short pulses might not be crucial. If the perfect phase matching condition is assumed, the only remaining mechanisms to decrease efficiency are self and cross phase modulation. Here we suggest that, instead of exactly matching wave vectors, we admit a small mismatch and show how it can be tailored to compensate for the cross phase modulation of the third harmonic by the fundamental during the generation process. This is very beneficial for the efficiency of third harmonic generation, even increasing it by a factor of two or more.

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As is well known, a monochromatic beam of frequency ω can generate a third harmonic, 3ω, in a nonlinear medium with a Kerr type response [1–4]. Even though application of this method as an effective technique to produce intense, tunable coherent light has somehow faded and been replaced by other efforts, it still remains one of the most fundamental phenomena in nonlinear optics. Moreover, recently third harmonic generation near the focal point of a tightly focused beam was used to probe the microscopical structure of transparent samples [5]. Possible modifications of these techniques in the high field regime were also suggested

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[6]. As was mentioned in [6,7], at higher intensities self and cross-phase modulation and even higher order effects can no longer be neglected in the description of the dynamics [8]. The role of the mismatch was mentioned in this context.

The efficiency of harmonic generation in ultrashort lasers is limited by the phase mismatch and temporal walk-off of the fundamental and harmonic pulses due to the phase and group velocity dispersion. Hopefully it will be possible to devise nonlinear optical media where the difference in group velocity between the fundamental and third harmonic is strongly suppressed [9]. Still a wavenumber mismatch will appear and we propose to put it to work so as to enhance third harmonic generation. A somewhat similar idea has been used to enhance the mixing of four de Broglie waves in a BEC [10]. The idea of using the mismatch δk in the process of pulse creation has also been the basis of soliton pair creation in [3,11]. It was demonstrated that a whole new, broad class of solitons exists for $\delta k \neq 0$.

Our model will be a nonlinear optical medium with a Kerr-type nonlinear susceptibility. We consider the resonant interaction between a linearly polarized beam of frequency ω and its third harmonic. The scalar wave equation for the propagating electric field is

$$\frac{\partial^2 E}{\partial z^2} + \nabla_\perp^2 E - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 (P_\mathrm{L} + P_\mathrm{NL})}{\partial t^2}. \tag{1}$$

Here the transverse laplacian is $\nabla_\perp^2 = \partial^2/\partial x^2$ for a slab waveguide and $1/r(\partial/\partial r)r(\partial/\partial r)$ for cylindrical symmetry; the latter case will be treated here. We consider the electric field for the fundamental and third harmonic,

$$E = \frac{1}{2}\{E_1\,\mathrm{e}^{\mathrm{i}(k_1 z - \omega t)} + E_3\,\mathrm{e}^{\mathrm{i}(k_3 z - 3\omega t)} + \mathrm{c.c.}\}, \tag{2}$$

where $k_j = j\omega n_j/c$, $n_j = n(jw)$, $j = 1, 3$. P_L is the linear polarization and P_NL is equal to

$$P_\mathrm{NL}(\omega) = \frac{\chi^{(3)}}{8}\Big[(3|E_1|^2 + 6|E_3|^2)E_1 + 3E_1^{*2}E_3\,\mathrm{e}^{-\mathrm{i}\delta k z}\Big]\mathrm{e}^{\mathrm{i}(k_1 z - \omega t)}, \tag{3}$$

where $\delta k = 3k_1 - k_3$ and a similar response for $P_\mathrm{NL}(3\omega)$. When these values are inserted into (1), we obtain two coupled NLS equations for E_1 and E_3:

$$2\mathrm{i}k_1\left(\frac{\partial E_1}{\partial z} + \frac{1}{v_1}\frac{\partial E_1}{\partial t}\right) + \nabla_\perp^2 E_1 = -\chi(|E_1|^2 + 2|E_3|^2)E_1 - \chi(E_1^*)^2 E_3\,\mathrm{e}^{-\mathrm{i}\delta k z}, \tag{4}$$

$$2\mathrm{i}k_3\left(\frac{\partial E_3}{\partial z} + \frac{1}{v_3}\frac{\partial E_3}{\partial t}\right) + \nabla_\perp^2 E_3 = -9\chi(|E_3|^2 + 2|E_1|^2)E_3 - 3\chi E_1^3\,\mathrm{e}^{\mathrm{i}\delta k z}, \tag{5}$$

where χ is $3\pi(\omega/c)^2\chi^{(3)}$ and is positive for a self-focusing and negative for a self-defocusing medium. Here v_1 and v_3 are the group velocities of the fundamental and third harmonic respectively. Suppose for the moment that the perpendicular dependence can be neglected, as being of higher order in Eqs. (4) and (5). We wish to see how E_3 can be most effectively built up from zero, working in the $|E_3| \ll |E_1|$ regime. We will first consider the case $v_1 = v_3 = v$. This case is important, as differences in these two group velocities can hopefully be suppressed. We will perform calculations in the coordinate system of the moving wavepacket's group velocities. The first equation (4) can then be approximated by

$$2\mathrm{i}k_1\frac{\partial E_1}{\partial z} = -\chi|E_1|^2 E_1, \tag{6}$$

and solved (r is the radial cylindrical variable):

$$E_1(z,r,t) = E_1(0,r,t)\mathrm{e}^{\mathrm{i}(\chi|E_1(0,r,t)|^2/2k_1)z}. \tag{7}$$

The terms we retain from the E_3 equation, (5), are

$$2\mathrm{i}k_3\frac{\partial E_3}{\partial z} = -18\chi|E_1|^2 E_3 - 3\chi E_1^3\,\mathrm{e}^{\mathrm{i}\delta k z}. \tag{8}$$

When this equation is solved and $|E_3|^2$ calculated, we find

$$|E_3(z,r,t)|^2 = \frac{9\chi^2}{4k_3^2}|E_1(0,r,t)|^6\frac{\sin^2(\Delta z)}{\Delta^2} \tag{9}$$

with

$$\Delta = \frac{\delta k}{2} - \frac{\chi E_1^2(0,r,t)}{2}\left(\frac{9}{k_3} - \frac{3}{2k_1}\right). \tag{10}$$

To calculate the optimal value of δk we must calculate the integral

$$\int |E_3(z,r,t)|^2 r\,\mathrm{d}r\,\mathrm{d}t = \frac{9\chi^2}{4k_3^2}\int |E_1(0,r,t)|^6 \frac{\sin^2(\Delta z)}{\Delta^2} r\,\mathrm{d}r\,\mathrm{d}t. \quad (11)$$

For small values of $|E_1(0,r,t)|$ we can calculate the integral (11) with accuracy up to second order upon taking $\sin(\Delta z) \approx \Delta z - (1/6)(\Delta z)^3$, where Δ is given by Eq. (10). We then find the optimal value of δk, for which the variation of the integral (11) is zero and check that it corresponds to a maximum. We find

$$\frac{\delta k}{k_1} = \alpha\left[\frac{\chi E_0^2}{k_1^2}\right] + \mathrm{O}\left[\frac{\chi E_0^2}{k_1^2}\right]^2, \quad (12)$$

where $E_0 \equiv E_1(0,0,0)$, and α depends on the assumed profile of $E_1(0,r,t)$. We adjust δk to maximize the integral (11) for various pulse shapes. Fig. 1 shows the value of the normalized δk defined as $\delta k \cdot k_1/(\chi E_0^2)$, as a function of $\chi E_0^2/k_1^2$, where E_0 is the maximum of the initial pulse strength. We consider four different initial pulse profiles for the fundamental wave: $E_1(0,r,t) = E_0 f(r)g(t)$, where $f(r)$ and $g(t)$ are: (a) square distribution functions, (b) parabolic distribution functions, (c) Gaussian distribution function and (d) Lorentzian distribution function, all $f(0) = g(0) = 1$. The values of α are respectively: 1.50, 1.02, 0.98, 0.91. Importantly, these values do not depend on the width of the model functions $f(r)$ and $g(t)$. For example if $f(r) = \exp(-[r/\sigma]^2)$, the value of σ will not matter. Even if the waist of the beam gradually narrows towards a focus, σ will be a slowly varying function of z; e.g. $\sigma^2 = \sigma_0^2(1+\epsilon z^2)$, but our result will not be effected. Numerics confirm this.

Fig. 2 shows the fraction of the total energy in the third harmonic $\int |E_3(z,r,t)|^2 r\,\mathrm{d}r\,\mathrm{d}t$ as a function of δk for different input energies. We kept

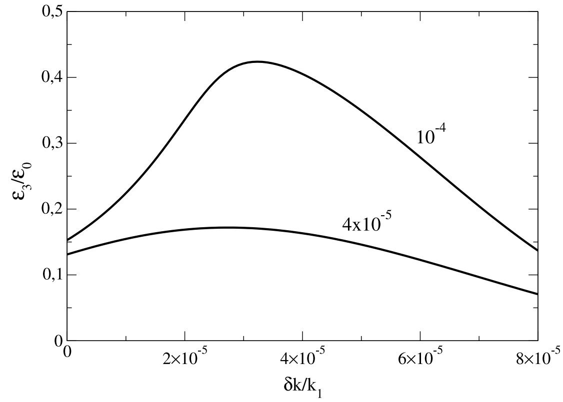

Fig. 2. Dependence of the fraction of the total energy ε_3 in the third harmonic as a function of our phase mismatch $\delta k/k_1 = 3 - k_3/k_1$. Here ε_0 is the initial energy in the fundamental wave, $z = 5$ mm and the two curves correspond to values of $\chi E_0^2/k_1^2$ as indicated.

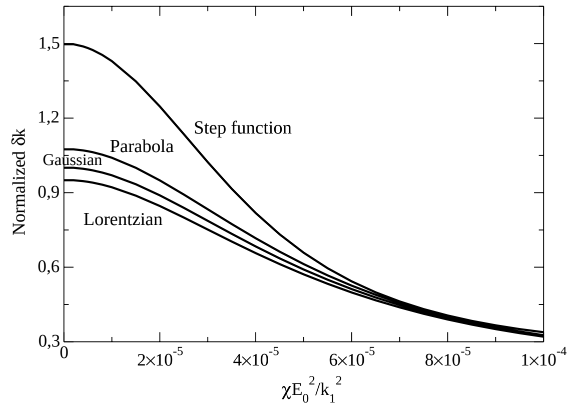

Fig. 1. Normalized phase mismatch δk as a function of $\chi E_0^2/k_1^2$. The propagation distance $z = 5$ mm ($\lambda = 0.8$ μm). The limits for E_0 tending to zero calculated from Eq. (11) agree very well with the numerically found values. (The maximal value of $\chi(E_0)^2/k_1^2, 1\times10^{-4}$, corresponds to the energy of 32 μJ for $n_2 = 2.8\times10^{-20}$ m²/W in fused silica.)

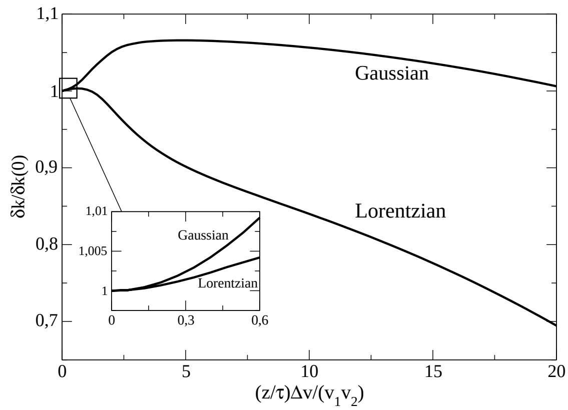

Fig. 3. Dependence of optimal phase mismatch δk on Δv (group velocity mismatch) for two initial profiles (c) and (d), $z = 5$ mm and the value of χE_0^2, was taken in the weak nonlinear coupling regime.

$\Delta v = 0$ and used Gaussians for both f and g (case (c)). The maxima of the third harmonic occur at values of $\delta k \neq 0$ and can be more than twice as large as for perfect phase matching. As we see from this figure the optimal value of δk is larger and better pronounced for larger intensities of the fundamental beam.

When $v_1 - v_3 = \Delta v > 0$, corrections to δk can be shown to be initially quadratic in Δv, as can be seen in Fig. 3. This figure shows the dependence of δk on Δv for the most physical initial $E_1(0, r, t)$ profiles, namely (c) and (d). For small group velocity mismatch the dependence is quadratic in both cases, as follows from symmetry. However, it

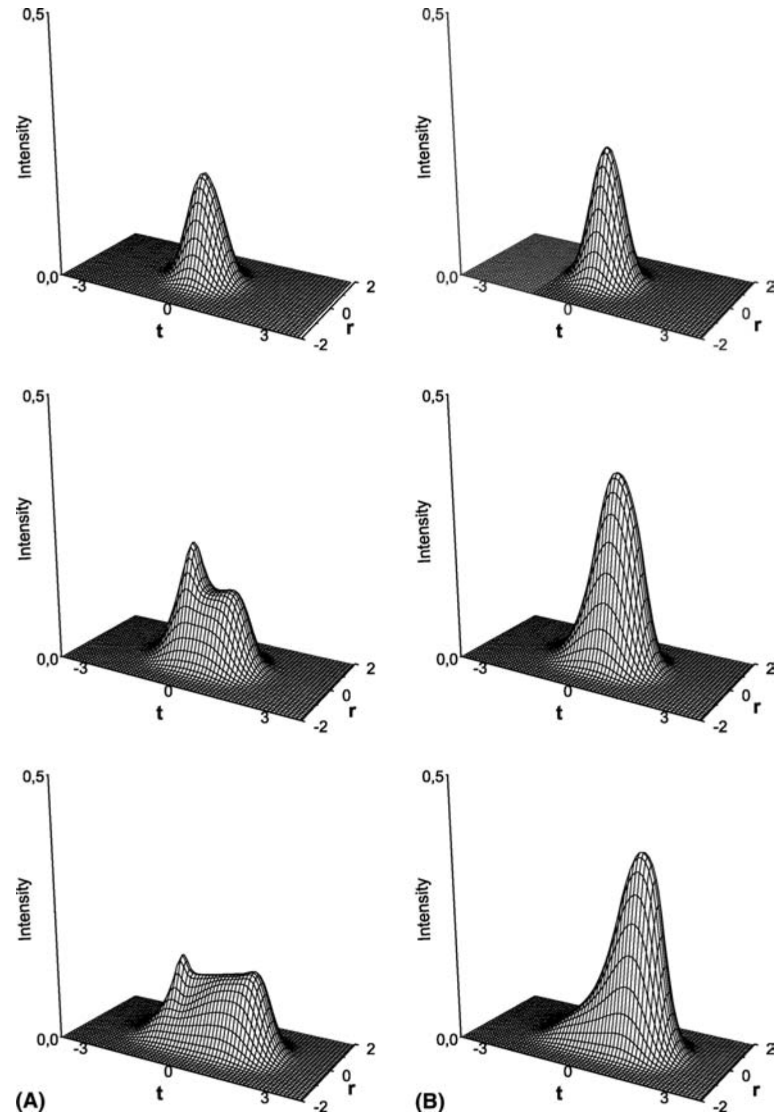

Fig. 4. Three dimensional plot of the intensity, $|E_3/E_0|^2$ of the third harmonic for increasing values of the propagation distance $z = 1.2$ mm, $z = 3$ mm, $z = 4.8$ mm for: (A) no mismatch and (B) optimal phase mismatch δk. The initial profile of the fundamental wave was $E_0\,e^{-[(r/\sigma)^2+(t/\tau)^2]}$, where $\sigma = 150$ μm and $\tau = 100$ fs. Here t is in units of τ, and r in units of σ, $\chi E_0^2/k_1^2 = 10^{-4}$ and $(z/\tau)\Delta v/(v_1 v_3) = 4$.

becomes substantially different for larger values of Δv.

Fig. 4 shows the development of the third harmonic for various propagation distances z (A) for perfect phase matching and (B) with mismatch δk as calculated above. Once again, we see that the intensity can be more than doubled by engineering a proper mismatch.

In conclusion, we have shown that a small mismatch can be beneficial for third harmonic generation. We derived a variational principle yielding optimal values of δk and found numerical values of δk for various initial pulse shapes. In the weak E_0 limit they agreed with theoretically found values. We also discussed the role of diffraction and group velocity dispersion. Finally, three dimensional plots of the third harmonic generation illustrate the difference in the dynamics between perfectly phase matched and optimal δk cases. Here too the phase mismatch more than doubles the efficiency of third harmonic generation.

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