Third-harmonic generation in isotropic media by focused pulses

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For focused pulses of light in isotropic nonlinear media, third-harmonic generation can be strongly affected by group-velocity mismatch between the fundamental and third-harmonic. There is a characteristic time determined by the group-velocity mismatch and the Rayleigh range of the focused pulse. The dynamics depend on two dimensionless quantities, namely the ratio of the characteristic time to the pulse duration and the phase-velocity mismatch times the Rayleigh range. Pulses shorter than the characteristic time have physics described by simple analytic formulas. Pulses near the characteristic time have an intermediate behavior given by an explicit but more complicated formula. Pulses longer than the characteristic time tend to the continuous-wave case.

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I. INTRODUCTION

Third-harmonic generation (THG) by focused *pulses* of light may, for sufficiently long pulses, be accurately described by a continuous wave (cw) beam model [1–4]. We present a quantitative analysis of the limits of the validity of the cw approximation for THG by focused pulses, and elaborate the physics in regimes where the cw approximation fails. For the instances of THG by focused pulses simulated numerically in Ref. [5], the group-velocity mismatch between the fundamental and third-harmonic (TH) was the most significant source of divergence from the conventional cw beam model [1,2]; Kerr and Raman effects, and higher-order dispersion and diffraction, were much weaker perturbations. Consequently, we formulate a model for THG by a pulse, including only the most essential effects: phase-velocity mismatch, group-velocity mismatch, diffraction, and THG. Omission of Kerr effects and other nonlinearities makes the problem much more tractable, even allowing analytic solutions. Two dimensionless parameters determine the nature of the dynamics: the first is a function of the group-velocity mismatch, the Rayleigh range (or the tightness of the focusing), and the pulse duration; the second is the phase-velocity mismatch times the Rayleigh range. If the first parameter is small (short pulses) or large (long pulses), the results are especially simple. In the intermediate case, the results are more complex, though still analytic. We apply the same analysis to higher-harmonic generation.

Harmonic generation is one of the basic nonlinear optical effects, and is the subject of a great deal of study. Second-harmonic generation (SHG), which requires a material without parity inversion symmetry, has garnered more attention than THG, because it is a lower-order effect. The basic SHG dynamics in waveguides, and also Kerr and other perturbations, may be found in Ref. [6], and citations therein. Exact solutions for SHG by Gaussian (i.e., focused) cw beams have been known since 1966 [2,7]. Work on SHG by focused *pulses*, including some exact solutions, may be found in Ref. [8] and citations therein. Solutions to THG by focused cw beams [1,2] were found a few years after those of SHG. The literature on THG in waveguides, and for effective plane waves in crystals, for which diffraction is unimportant, is too extensive to summarize easily here. Some notable work on THG related to that presented here is as follows. Reference [9] finds group-velocity mismatch effects on THG in a fiber, and also includes phase-velocity mismatch and Kerr effects; Ref. [10] considers spectral broadening of THG pulses resulting from phase-velocity mismatch and Kerr effects; Ref. [11] examines the consequences of Kerr effects on THG spectra without any linear mismatch. Reference [12] covers effective THG via cascaded SHG, including phase- and group-velocity mismatch, dispersion, and Kerr effects; Ref. [13] looks at effective THG by cascaded SHG by focused pulses, including phase-velocity mismatch, Kerr effects, and parametric down-conversion; Ref. [14] deals with THG via cascaded SHG, with phase-velocity matching, group-velocity mismatch, and dispersion. To our knowledge, effects of group-velocity mismatch on THG by focused pulses (i.e., where diffraction is important, and where the cw approximation is insufficient) have not been studied.

II. MODEL

Equations that minimally describe pulses of light comprised of a fundamental frequency and its TH in a bulk isotropic medium are

$$0 = i\frac{\partial}{\partial z}u_1 + ik_1'\frac{\partial}{\partial t}u_1 + \frac{1}{2k_1}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)u_1 + \frac{2\pi(\omega_1/c)^2}{k_1} \times \exp[-i(3k_1 - k_3)z]\chi^{\mathrm{THG}}3u_1^{*2}u_3, \tag{1a}$$

$$0 = i\frac{\partial}{\partial z}u_3 + ik_3'\frac{\partial}{\partial t}u_3 + \frac{1}{2k_3}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)u_3 + \frac{2\pi(3\omega_1/c)^2}{k_3}\exp[i(3k_1 - k_3)z]\chi^{\mathrm{THG}}(u_1)^3, \tag{1b}$$

where $u_1(t,x,y,z)$ is the fundamental slowly varying envelope, with carrier wave $\exp[i(k_1 z - \omega_1 t)]$, and $u_3(t,x,y,z)$ is the envelope of the TH, with carrier wave $\exp[i(k_3 z - 3\omega_1 t)]$. The respective wave numbers are $k_1 \equiv n(\omega_1)\omega_1/c$ and $k_3 \equiv n(3\omega_1)3\omega_1/c$, and the reciprocal group velocities are $k_1' \equiv (d/d\omega)[n(\omega)\omega/c]_{\omega=\omega_1}$ and $k_3' \equiv (d/d\omega)[n(\omega)\omega/c]_{\omega=3\omega_1}$, with $n(\omega)$ the index of refraction. The second derivatives with respect to x and y represent (lowest-order) diffraction. THG is taken as effectively instantaneous. Dispersion, higher-order diffraction, and Kerr and other nonlinear effects are neglected. Equations (1) are appropriate for paraxial pulses that are not so intense that Kerr effects become important, for which the percentage of the energy in the third-harmonic band remains relatively small, and for which dispersion is relatively unimportant. When a relatively small part of the energy is in the TH, the rightmost term in Eq. (1a)—a $\chi^{(3)}$ form of parametric down-conversion, or a downshifting counterpart to THG—may be dropped (making the equations non-Hamiltonian), which we do henceforth. Equation (1a) has an exact solution with a spatially Gaussian profile, and an arbitrary temporal profile translating at group velocity $(1/k_1')$,

$$u_1(t,x,y,z) = f(t - k_1'z)\frac{\sqrt{k_1 z_R/\pi}}{z_R + iz}\exp\left(-\frac{k_1}{z_R + iz}\frac{x^2+y^2}{2}\right). \tag{2a}$$

We assume that the temporal profile is Gaussian,

$$f(t) = \frac{\sqrt{E_1/\sqrt{\pi}}}{t_p}\exp\left(-\frac{1}{2}\frac{t^2}{t_p^2}\right). \tag{2b}$$

The coefficients in Eqs. (2) give normalizion $\int\int\int|u_1|^2 dtdxdy = E_1$, which is proportional to the pulse energy. Without loss of generality, the Rayleigh range (or half the confocal parameter), z_R, is taken to be positive, and the phase of the fundamental is chosen to be zero at the focus $z=0$.

III. SOLUTION FOR THE THIRD-HARMONIC PULSE

To convert the problem from a partial differential equation to an ordinary differential equation, insert the fundamental pulse (2) into the equation for the TH (1b), and Fourier-transform

$$f(\omega,k_x,k_y) = \mathcal{F}\{f(t,x,y)\} \equiv (2\pi)^{-3/2}\int\int\int_{-\infty}^{\infty} f(t;x,y)\exp[i(\omega t - k_x x - k_y y)]dtdxdy$$

from real space to momentum space,

$$0 = i\frac{\partial}{\partial z}u_3(\omega,k_x,k_y,z) + k_3'\omega u_3 - \frac{k_x^2+k_y^2}{2k_3}u_3 + \frac{2\pi(3\omega_1/c)^2}{k_3}\chi^{\mathrm{THG}}\exp[i(3k_1-k_3)z]\mathcal{F}\{u_1(t,x,y,z)^3\} \tag{3a}$$

$$= i\frac{\partial}{\partial z}u_3(\omega,k_x,k_y,z) + \left(k_3'\omega - \frac{k_x^2+k_y^2}{2k_3}\right)u_3 + \chi^{\mathrm{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\frac{2\sqrt{3}}{\pi^{5/4}}\frac{(k_1 z_R E_1)^{3/2}/t_{\mathrm{pulse}}^2}{(z_R+iz)^2} \times\exp\left[i(3k_1 - k_3 + k_1'\omega)z - \frac{t_{\mathrm{pulse}}^2}{3}\frac{\omega^2}{2} - \frac{z_R+iz}{3k_1}\frac{k_x^2+k_y^2}{2}\right]. \tag{3b}$$

This inhomogeneous first-order linear differential equation can be integrated. We choose a constant of integration such that the TH field is zero at $z=z_0$. Physically, this describes a fundamental pulse incident on a nonlinear medium at distance $z=z_0$. The result is a solution for the TH pulse in momentum space (ω,k_x,k_y,z); it is also useful to have the solution inverse Fourier-transformed to real space,

$$u_3(\omega,k_x,k_y,z) = i\chi^{\mathrm{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\frac{2}{\pi^{5/4}}\frac{t_p}{\sqrt{3}}\left(\frac{k_1 z_R E_1}{t_p^2}\right)^{3/2}\exp\left[-\frac{t_p^2}{3}\frac{\omega^2}{2} - \frac{z_R}{3k_1}\frac{k_x^2+k_y^2}{2} + i\left(k_3'\omega - \frac{k_x^2+k_y^2}{2k_3}\right)z\right] \times\int_{z_0}^{z}\frac{1}{(z_R+iz')^2}\exp\left\{i\left[(3k_1-k_3)\left(1+\frac{k_x^2+k_y^2}{2(3k_1)k_3}\right) + (k_1'-k_3')\omega\right]z'\right\}dz', \tag{4a}$$

$$u_3(\omega,x,y,z) = i\chi^{\mathrm{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\frac{2}{\pi^{5/4}}\frac{t_p}{\sqrt{3}}\left(\frac{k_1 z_R E_1}{t_p^2}\right)^{3/2}\exp\left(-\frac{\omega^2 t_p^2}{6} + ik_3'\omega z\right) \times\int_{z_0}^{z}\frac{1}{(z_R+iz')^2}\left(\frac{z_R}{3k_1} + i\frac{z}{k_3} - i\frac{3k_1-k_3}{3k_1k_3}z'\right)^{-1} \times\exp\left[-\left(\frac{z_R}{3k_1} + i\frac{z}{k_3} - i\frac{3k_1-k_3}{3k_1k_3}z'\right)^{-1}\frac{x^2+y^2}{2} + i(3k_1 - k_3 + (k_1'-k_3')\omega)z'\right]dz', \tag{4b}$$

$$u_3(t,k_x,k_y,z) = i\chi^{\mathrm{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\frac{2}{\pi^{5/4}}\left(\frac{k_1z_RE_1}{t_p^2}\right)^{3/2}\exp\left[-\left(\frac{z_R}{3k_1}+i\frac{z}{k_3}\right)\frac{k_x^2+k_y^2}{2}\right]$$
$$\times\int_{z_0}^{z}\frac{1}{(z_R+iz')^2}\exp\left[-\frac{3}{2}\left(\frac{t-k_3'z-(k_1'-k_3')z'}{t_p}\right)^2+i(3k_1-k_3)\left(1+\frac{k_x^2+k_y^2}{2(3k_1)k_3}\right)z'\right]dz', \tag{4c}$$

$$u_3(t,x,y,z) = i\chi^{\mathrm{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\frac{2}{\pi^{5/4}}\left(\frac{k_1z_RE_1}{t_p^2}\right)^{3/2}\int_{z_0}^{z}\frac{dz'}{(z_R+iz')^2}\left(\frac{z_R}{3k_1}+i\frac{z}{k_3}-i\frac{3k_1-k_3}{3k_1k_3}z'\right)^{-1}$$
$$\times\exp\left[-\frac{3}{2}\left(\frac{t-k_3'z-(k_1'-k_3')z'}{t_p}\right)^2-\left(\frac{z_R}{3k_1}+i\frac{z}{k_3}-i\frac{3k_1-k_3}{3k_1k_3}z'\right)^{-1}\frac{x^2+y^2}{2}+i(3k_1-k_3)z'\right]. \tag{4d}$$

All forms (4) of the solution are equivalent. This is the general and mathematically exact solution for a TH field due to a fundamental pulse of the form (2) in Eqs. (1) without the parametric down-conversion term.

IV. ANALYSIS

The physics may be elucidated by examining some limits and approximations. The TH far field is, in general, not an exact Gaussian, either in time or in space. The pulse takes a temporally Gaussian form in the case of perfect group-velocity matching, $k_1'=k_3'$. The pulse takes a spatially Gaussian form in the case of perfect phase-velocity matching, $3k_1=k_3$. Otherwise, with a nonvanishing phase-velocity mismatch, the radial momentum (k_x,k_y) in the integrand gives the TH field a more complex form. For many purposes, however, the pulse is very close to being spatially Gaussian, and we may make the approximation $(3k_1-k_3)(k_x^2+k_y^2)\to 0$. Indeed, it is conventional to do so [1,2]. In this approximation, the TH field can be expressed in real space and/or momentum space,

$$u_3(\omega,k_x,k_y,z) = i\chi^{\mathrm{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\frac{2}{\pi^{5/4}}\frac{t_p}{\sqrt{3}}\left(\frac{k_1z_RE_1}{t_p^2}\right)^{3/2}\exp\left[-\frac{t_p^2}{3}\frac{\omega^2}{2}-\frac{z_R}{3k_1}\frac{k_x^2+k_y^2}{2}+i\left(k_3'\omega-\frac{k_x^2+k_y^2}{2k_3}\right)z\right]$$
$$\times\int_{z_0}^{z}\frac{1}{(z_R+iz')^2}\exp\{i[3k_1-k_3+(k_1'-k_3')\omega]z'\}dz', \tag{5a}$$

$$u_3(\omega,x,y,z) = i\chi^{\mathrm{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\frac{2}{\pi^{5/4}}\frac{t_p}{\sqrt{3}}\left(\frac{k_1z_RE_1}{t_p^2}\right)^{3/2}\left(\frac{z_R}{3k_1}+i\frac{z}{k_3}\right)^{-1}\exp\left[-\frac{\omega^2t_p^2}{6}-\left(\frac{z_R}{3k_1}+i\frac{z}{k_3}\right)^{-1}\frac{x^2+y^2}{2}+ik_3'\omega z\right]$$
$$\times\int_{z_0}^{z}\frac{1}{(z_R+iz')^2}\exp\{i[3k_1-k_3+(k_1'-k_3')\omega]z'\}dz', \tag{5b}$$

$$u_3(t,k_x,k_y,z) = i\chi^{\mathrm{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\frac{2}{\pi^{5/4}}\left(\frac{k_1z_RE_1}{t_p^2}\right)^{3/2}\exp\left[-\left(\frac{z_R}{3k_1}+i\frac{z}{k_3}\right)\frac{k_x^2+k_y^2}{2}\right]$$
$$\times\int_{z_0}^{z}\frac{1}{(z_R+iz')^2}\exp\left[-\frac{3}{2}\left(\frac{t-k_3'z-(k_1'-k_3')z'}{t_p}\right)^2+i(3k_1-k_3)z'\right]dz', \tag{5c}$$

$$u_3(t,x,y,z) = i\chi^{\mathrm{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\frac{2}{\pi^{5/4}}\left(\frac{k_1z_RE_1}{t_p^2}\right)^{3/2}\left(\frac{z_R}{3k_1}+i\frac{z}{k_3}\right)^{-1}\exp\left[-\left(\frac{z_R}{3k_1}+i\frac{z}{k_3}\right)^{-1}\frac{x^2+y^2}{2}\right]$$
$$\times\int_{z_0}^{z}\frac{1}{(z_R+iz')^2}\exp\left[-\frac{3}{2}\left(\frac{t-k_3'z-(k_1'-k_3')z'}{t_p}\right)^2+i(3k_1-k_3)z'\right]dz'. \tag{5d}$$

Three limits have analytic solutions. One is propagation from a near field to a near field [2]. Here, diffraction plays no important part, and the pulse approaches a plane wave. In this case, none of the methodology developed here is relevant. Second is the limit in which the pulse starts and ends far away from the focus, or far-field to far-field propagation. Here, integrals may be considered as starting and ending at infinity, and they may be analytically integrated. Third is propagation from a far-field to a focus, or vice versa. In this limit, the integrals may be transformed into special functions.

A. Far-field to far-field propagation

In the limit of far-field to far-field propagation ($z_0 \ll -z_R$ and $z \gg z_R$), the integral (4a) may be solved because the integrand is analytic except for a second-order pole at $z = iz_R$,

$$u_3(\omega,k_x,k_y,z) = i\chi^{\text{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\frac{4}{\pi^{1/4}}\frac{t_p/\sqrt{3}}{z_R}\left(\frac{k_1 z_R E_1}{t_p^2}\right)^{3/2} \times \phi \exp\left[-\phi - \frac{t_p^2}{3}\frac{\omega^2}{2} - \frac{z_R}{3k_1}\frac{k_x^2+k_y^2}{2} + i\left(k_3'\omega - \frac{k_x^2+k_y^2}{2k_3}\right)z\right]H(\phi), \tag{6a}$$

with generalized dimensionless mismatch going to

$$\phi = \phi(\omega,k_x,k_y) \equiv \left[(3k_1 - k_3)\left(1 + \frac{k_x^2+k_y^2}{2(3k_1)k_3}\right) + (k_1' - k_3')\omega\right]z_R, \tag{6b}$$

and H is the step function,

$$H(x) = \begin{cases} 1, & x \geq 0 \\ 0, & x < 0. \end{cases} \tag{6c}$$

In the far-field limit, when the TH field is expressed as a function of momentum space (ω,k_x,k_y), there is a z dependence in the phase, but not in the magnitude. The TH pulse thus continues to propagate—with group velocity $1/k_3'$—and to diverge, but in an essentially trivial manner. Dependence of the TH power on the dimensionless mismatch ϕ is shown in Fig. 1. The spectrum vanishes at negative ϕ, peaks at $\phi = 1$, and drops off at large ϕ. The nontrivial dynamics occur only in the near or intermediate field. Real-space expressions are not available in as simple a form because the contour does not vanish at large imaginary values of z', or the function is not meromorphic in real space.

If we neglect spatially non-Gaussian features $[(3k_1 - k_3)(k_x^2+k_y^2) \to 0]$, the TH field is given by Eq. (6a), where the dimensionless mismatch parameter is

$$\phi = \left(1 + \frac{k_1' - k_3'}{3k_1 - k_3}\omega\right)(3k_1 - k_3)z_R. \tag{7}$$

Expressions in terms of time rather than frequency may be obtained, but not simplified in the same way, since the integrand does not vanish at large imaginary values. Also, the complexity of these forms make them less useful. The power spectrum of the TH band is obtained by integrating the intensity over transverse coordinates,

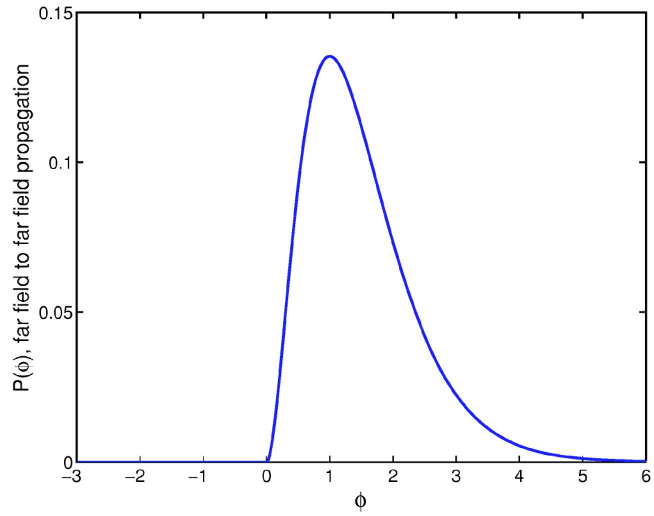

FIG. 1. Power spectrum of the TH pulse, after far-field to far-field propagation: dependence on the dimensionless mismatch $\phi = \{(3k_1 - k_3)[1 + (k_x^2+k_y^2)/(6k_1k_3)] + (k_1' - k_3')\omega\}z_R$, holding other parameters constant, $P \propto \phi^2 \exp(-2\phi)H(\phi)$. Here, k_1 and k_3 are the wave vectors in the fundamental and TH, k_1' and k_3' are their derivatives with respect to frequency, ω is frequency within the TH, k_x and k_y are transverse wave numbers, z_R is the Rayleigh range, and H is the step function.

$$P(3\omega_1 + \omega) = \int\int |u_3(\omega,k_x,k_y,z)|^2 dk_x dk_y = \int\int |u_3(\omega,x,y,z)|^2 dx dy = \left[\chi^{\text{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\right]^2 16\sqrt{\pi}\frac{k_1^4E_1^3}{t_p^4}\phi^2 \times \exp\left(-2\phi - \frac{\omega^2 t_p^2}{3}\right)H(\phi), \tag{8}$$

where ϕ is given by Eq. (7).

Three parameters determine the *qualitative* properties of the TH output: the fundamental pulse width t_p, the characteristic time scale

$$t_{\text{char}} \equiv (k_1' - k_3')z_R, \tag{9}$$

which can be either positive or negative, and the phase-velocity mismatch times the Rayleigh range, $(3k_1 - k_3)z_R$, which is dimensionless. The ratio of the first two parameters is dimensionless, and it, together with the third parameter, are the two dimensionless parameters that determine the dynamics.

1. Short duration pulses

When the fundamental pulse is much shorter than the magnitude of the characteristic time scale, $t_p \ll |t_{\text{char}}| = |k_1' - k_3'|z_R$, the power spectrum in the region of the TH is essentially determined by the medium and the Rayleigh range of the pulse, while the fundamental pulse width only scales the entire TH spectrum by a factor. The peak spectral density in the TH band is then

$$P_{\text{peak}}^{\text{THG}} = \left[\chi^{\text{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\right]^2\frac{16\sqrt{\pi}}{e^2}\exp\left[-\frac{t_p^2(3k_1-k_3)^2}{6(k_1'-k_3')^2}\right]\frac{k_1^4E_1^3}{t_p^4} \tag{10a}$$

at

$$\omega_{\text{peak}}^{\text{THG}} = 3\omega_1 - \frac{3k_1-k_3}{k_1'-k_3'}. \tag{10b}$$

The spectral width of the TH band is approximately given by

$$\Delta\omega^{\text{THG}} = \frac{1}{|k_1'-k_3'|z_R}, \tag{10c}$$

and the temporal width of the TH pulse is

$$\Delta t^{\text{THG}} = |k_1'-k_3'|z_R. \tag{10d}$$

The energy in the TH pulse is the integral over the TH power spectrum,

$$E_3 = \int P(3\omega_1+\omega)d\omega = \left[\chi^{\text{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\right]^2 4\sqrt{\pi} \times\exp\left[-\frac{t_p^2(3k_1-k_3)^2}{6(k_1'-k_3')^2}\right]\frac{k_1^4E_1^3}{t_p^4|k_1'-k_3'|z_R}. \tag{10e}$$

The assumption that dispersion is unimportant will break down for sufficiently short pulses. The results in this subsection hold for pulses that are short compared to the characteristic time of the focused pulse in the medium, $|k_1'-k_3'|z_R$, but not so short that the assumption herein that dispersion is relatively unimportant becomes inappropriate.

2. Long duration pulses

When the fundamental pulse is much longer than the magnitude of the characteristic time, $t_p \gg |t_{\text{char}}| = |k_1'-k_3'|z_R$, and the phase-velocity mismatch is positive, $(3k_1-k_3)z_R > 0$, the reverse holds: the TH power spectrum depends essentially on the fundamental pulse, and the properties of the medium merely scale the entire spectrum up or down by a factor. The peak spectral density is

$$P_{\text{peak}}^{\text{THG}} = \left[\chi^{\text{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\right]^2 16\sqrt{\pi}\frac{k_1^4E_1^3}{t_p^4}(3k_1-k_3)^2z_R^2 \times\exp[-2(3k_1-k_3)z_R], \tag{11a}$$

at, simply, triple the fundamental frequency,

$$\omega_{\text{peak}}^{\text{THG}} = 3\omega_1. \tag{11b}$$

In the long-pulse limit, the TH power spectrum approaches a simple Gaussian. The spectral width of the TH pulse is approximately

$$\Delta\omega^{\text{THG}} = \sqrt{3}/t_p, \tag{11c}$$

and the temporal width of the TH pulse is

$$\Delta t^{\text{THG}} = t_p/\sqrt{3}. \tag{11d}$$

The TH pulse energy is obtained by integrating the power spectrum over the TH band,

$$E_3 = \int P(3\omega_1+\omega)d\omega = \left[\chi^{\text{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\right]^2 16\sqrt{3}\pi\frac{k_1^4E_1^3}{t_p^5} \times(3k_1-k_3)^2z_R^2\exp[-2(3k_1-k_3)z_R]. \tag{11e}$$

Perfect group-velocity matching, $k_1'=k_3'$, is subsumed into this limit: the physics is like that of cw beams, no matter how short the pulses are.

When the fundamental pulse is long compared to the magnitude of the characteristic time scale, $t_p \gg |t_{\text{char}}|$, but the phase-velocity mismatch is negative, $(3k_1-k_3)z_R < 0$, the far-field TH energy comes only from the tail of the fundamental pulse's frequency distribution. This is because the step function in the power spectrum (8) totally cuts off light near the center of the TH band (leaving very little TH energy). The spectral peak is at

$$\omega_{\text{peak}}^{\text{THG}} = 3\omega_1 + \left[\frac{1}{4} - (3k_1-k_3)z_R\right]\frac{k_1'-k_3'}{3k_1-k_3}\frac{3}{t_p^2} + O(t_p^{-4}). \tag{12}$$

The TH spectral width and maximum are found by inserting this into the formula for the power spectrum (8). The TH that reaches the far-field, in this case of negative phase-velocity mismatch, shows a more pronounced influence due to the dimensionless mismatch (interference effects) than due to the fundamental power spectrum.

3. Intermediate duration pulses

When the fundamental pulse is of the same order as the characteristic time scale, the qualititive properties of the TH spectrum (8) may depend equally on the material and on the width of the fundamental pulse. In the general case, the peak spectral density occurs at

$$\omega_{\text{peak}}^{\text{THG}} = 3\omega_1 - \frac{1}{2}\left[\frac{3k_1-k_3}{k_1'-k_3'} + \frac{(k_1'-k_3')z_R}{t_p^2/3}\right] + \sqrt{\frac{1}{4}\left[\frac{3k_1-k_3}{k_1'-k_3'} - \frac{(k_1'-k_3')z_R}{t_p^2/3}\right]^2 + \frac{3}{t_p^2}}. \tag{13}$$

The spectral and temporal widths of the TH pulse are directly accessible from Eq. (8). The explicit forms are complex and opaque, so we do not write them out.

B. Focus to far-field propagation

The TH field that accumulates between a focus $(z=0)$ and a far-field $(z \gg z_R)$ can also be solved analytically, but only in the sense that the integral (4) transforms to an expression with special functions,

$$u_3(\omega,k_x,k_y,z) = \chi^{\mathrm{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\frac{2}{\pi^{5/4}}\frac{t_p/\sqrt{3}}{z_R}\left(\frac{k_1z_RE_1}{t_p^2}\right)^{3/2} \times\exp\left[-\frac{\omega^2t_p^2}{6}-\frac{z_R}{3k_1}\frac{k_x^2+k_y^2}{2} + i\left(k_3'\omega-\frac{k_x^2+k_y^2}{2k_3}\right)z\right] \times\left(1-\phi\frac{\mathrm{Ei}(\phi)+i\pi H(\phi)}{\exp(\phi)}\right), \quad (14a)$$

where the exponential integral is $\mathrm{Ei}(x)=-\int_{-x}^{\infty}t^{-1}\exp(-t)dt$, taking the principal value of the integral [15].

The net TH generated by propagation from a far-field to a focus is

$$u_3(\omega,k_x,k_y,0) = -\chi^{\mathrm{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\frac{2}{\pi^{5/4}}\frac{t_p/\sqrt{3}}{z_R}\left(\frac{k_1z_RE_1}{t_p^2}\right)^{3/2} \times\exp\left(-\frac{\omega^2t_p^2}{6}-\frac{z_R}{3k_1}\frac{k_x^2+k_y^2}{2}\right) \times\left(1-\phi\frac{\mathrm{Ei}(\phi)+i\pi H(\phi)}{\exp(\phi)}\right), \quad (14b)$$

Except for a difference in sign, and the fact that this far-field to focus result is expressed as the TH at the focus rather than approaching infinity, this result is similar to Eq. (14a), the focus to far-field result.

The power spectrum of the TH pulse, either to or from a focus, is

$$P(3\omega_1+\omega) = \int\int |u_3(\omega,k_x,k_y,z)|^2 dk_x dk_y = \left[\chi^{\mathrm{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\right]^2\frac{4}{\pi^{3/2}}\frac{k_1^4E_1^3}{t_p^4}\exp\left(-\frac{\omega^2t_p^2}{3}\right) \times\left|1-\phi\frac{\mathrm{Ei}(\phi)+i\pi H(\phi)}{\exp(\phi)}\right|^2. \quad (15)$$

Note the implicit frequency dependence via ϕ. Figure 2 shows the dependence of the TH power spectrum on the dimensionless mismatch ϕ. Neglecting the frequency dependence due to the finite width of the fundamental pulse, the maximum power occurs for mismatch $\phi_{\mathrm{peak}}\approx 0.437$ (in contrast to $\phi_{\mathrm{peak}}=1$ for the far-field to far-field limit). There is some spectral density at mismatches that are very large in either the positive or negative direction (in contrast to the complete cutoff for negative mismatch in the far-field to far-field limit), but the amount is relatively small for $\phi\lesssim -2$ or $\phi\gtrsim 6$.

1. Short-duration pulses

Let us neglect non-Gaussian spatial features, $(3k_1-k_3)(k_x^2+k_y^2)\to 0$. For very narrow pulses, $t_p\ll|k_1'-k_3'|z_R$, the spectral peak is

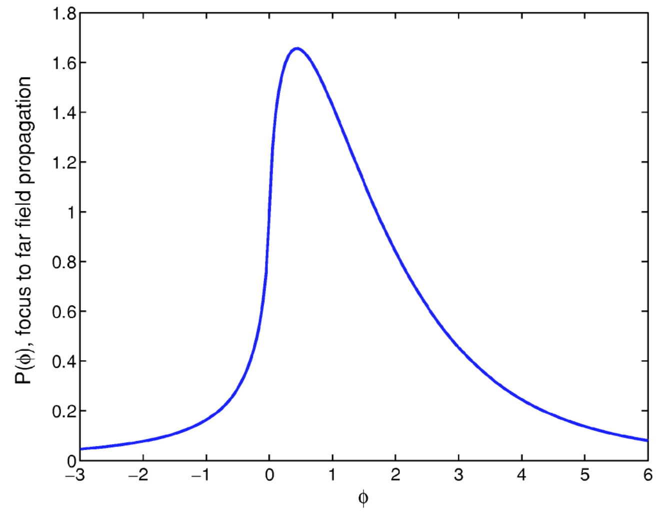

FIG. 2. Power spectrum of the TH pulse after focus to far-field propagation: dependence on the dimensionless mismatch $\phi=\{(3k_1-k_3)[1+(k_x^2+k_y^2)/(6k_1k_3)]+(k_1'-k_3')\omega\}z_R$, holding other parameters constant, $P\propto|1-\phi[\mathrm{Ei}(\phi)+i\pi H(\phi)]/\exp(\phi)|^2$. Here, k_1 and k_3 are the wave vectors in the fundamental and TH, k_1' and k_3' are their derivatives with respect to frequency, ω is frequency within the TH, k_x and k_y are transverse wave numbers, z_R is the Rayleigh range, Ei is the exponential integral, and H is the step function.

$$P_{\mathrm{peak}}^{\mathrm{THG}} \approx 1.19\left[\chi^{\mathrm{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\right]^2\exp\left[-\frac{t_p^2(3k_1-k_3)^2}{6(k_1'-k_3')^2}\right]\frac{k_1^4E_1^3}{t_p^4} \quad (16a)$$

at frequency

$$\omega_{\mathrm{peak}}^{\mathrm{THG}} \approx 3\omega_1 - 0.437\frac{3k_1-k_3}{k_1'-k_3'}. \quad (16b)$$

The spectral width is

$$\Delta\omega^{\mathrm{THG}} = \frac{2}{|k_1'-k_3'|z_R}, \quad (16c)$$

the temporal width is

$$\Delta t^{\mathrm{THG}} = |k_1'-k_3'|z_R/2, \quad (16d)$$

and the total energy in the TH pulse is

$$E_3 \approx 3.54\left[\chi^{\mathrm{THG}}\frac{(3\omega_1/c)^2}{3k_1k_3}\right]^2\exp\left[-\frac{t_p^2(3k_1-k_3)^2}{6(k_1'-k_3')^2}\right] \times\frac{k_1^4E_1^3}{t_p^4|k_1'-k_3'|z_R}. \quad (16e)$$

As in far-field to far-field propagation, these results hold for pulses that are short compared to the magnitude of the characteristic time of the focused pulse in the medium, $|k_1'-k_3'|z_R$, but not so short that dispersion becomes important.

2. Long-duration pulses

When the pulse is wide, $t_p\gg|k_1'-k_3'|z_R$, behavior is like in the cw limit. The spectral maximum is

$$P_{\text{peak}}^{\text{THG}} = \left[\chi^{\text{THG}} \frac{(3\omega_1/c)^2}{3k_1k_3} \right]^2 \frac{4}{\pi^{3/2}} \frac{k_1^4 E_1^3}{t_p^4} \left| 1 - (3k_1 - k_3) z_R \frac{\text{Ei}((3k_1-k_3)z_R) + i\pi H((3k_1-k_3)z_R)}{\exp[(3k_1-k_3)z_R]} \right|^2 \quad (17a)$$

at frequency

$$\omega_{\text{peak}}^{\text{THG}} \approx 3\omega_1. \quad (17b)$$

The spectral width is

$$\Delta\omega^{\text{THG}} = \sqrt{3}/t_p, \quad (17c)$$

the temporal width is

$$\Delta t^{\text{THG}} = t_p/\sqrt{3}, \quad (17d)$$

and the energy in the TH pulse is

$$E_3 = \left[\chi^{\text{THG}} \frac{(3\omega_1/c)^2}{3k_1k_3} \right]^2 \frac{4}{\sqrt{3}\pi} \frac{k_1^4 E_1^3}{t_p^3} \left| 1 - (3k_1 - k_3) z_R \times \frac{\text{Ei}((3k_1-k_3)z_R) + i\pi H((3k_1-k_3)z_R)}{\exp[(3k_1-k_3)z_R]} \right|^2. \quad (17e)$$

3. Intermediate duration pulses

The frequency dependence of the general power spectrum (15) comes from competition between two elements, namely (i) the Gaussian, which is due to the Gaussian spectrum of the fundamental pulse, and (ii) the function of the dimensionless phase mismatch ϕ, which is due to interference. The first element *tends* to maximize the spectrum at $\omega=0$ (within the TH band), which is the spectral peak for long duration pulses. The second element *tends* to maximize the spectrum at $\omega=-0.437(3k_1-k_3)/(k_1'-k_3')$, which is the spectral peak for short duration pulses. For short and for long pulses, one of the elements dominates. When the pulse is of intermediate duration, a small phase mismatch will put the two peaks close together, and they will merge. A large phase mismatch will put one of the peaks on the tail of the other, making the second tendency to peak insignificant. Figure 3 illustrates several intermediate instances, with pulse duration the same order as the characteristic time. The TH power spectrum is shown versus frequency, for a particular group-velocity mismatch and several phase-velocity mismatches.

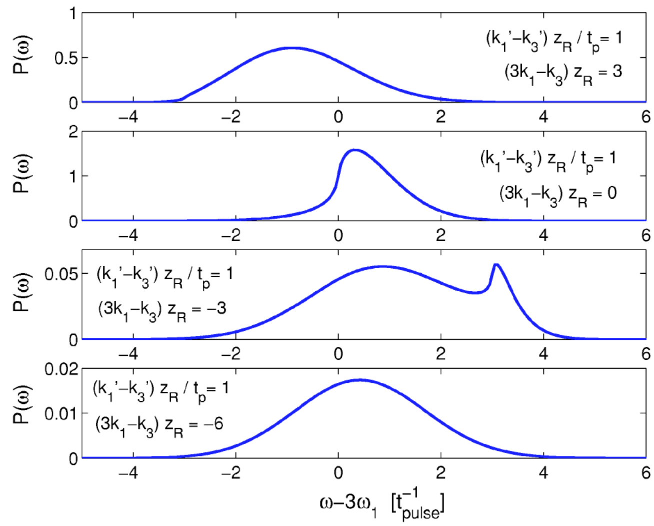

FIG. 3. Spectral density vs frequency, after focus to far-field propagation, with group-velocity mismatch $(k_1'-k_3')$ and Rayleigh range z_R such that the characteristic time scale is approximately equal to the pulse duration t_p, and for several values of the phase-velocity mismatch. The full frequency dependence of the power spectrum goes as $P \propto \exp(-t_p^2\omega^2/3)|1-\phi[\text{Ei}(\phi)+i\pi H(\phi)]/\exp(\phi)|^2$, with Ei the exponential integral and H the step function.

V. HIGHER-ORDER HARMONICS

The same analysis may be applied to a system with the nonlinearity of arbitrary-order harmonic generation. For two reasons, we will give the basic results, but we will not elaborate on the various limits and approximations. First, even-order susceptibilities require a medium with spatial inversion asymmetry. This is usually inconsistent with an isotropic optical medium. Second, lower-order processes in the electric field strength tend to be the most important in the most commonly encountered media and typical intensities. Thus THG is the most important instance of harmonic generation for isotropic media. The simplest governing equations for qth-order harmonic generation are

$$0 = i\frac{\partial}{\partial z}u_1 + ik_1'\frac{\partial}{\partial t}u_1 + \frac{1}{2k_1}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)u_1 + \frac{2\pi(\omega_1/c)^2}{k_1}\chi^{\text{qHG}} \exp[-i(qk_1-k_q)z]qu_1^{*2}u_q, \quad (18a)$$

$$0 = i\frac{\partial}{\partial z}u_q + ik_q'\frac{\partial}{\partial t}u_q + \frac{1}{2k_q}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)u_q + \frac{2\pi(q\omega_1/c)^2}{k_q}\chi^{\text{qHG}}\exp[i(qk_1-k_q)z](u_1)^q, \quad (18b)$$

where $u_q(t,x,y,z)$ is the envelope of carrier wave $\exp[i(k_q z - q\omega_1 t)]$, the wave number of the harmonic of order q is $k_q \equiv n(q\omega_1)q\omega_1/c$, and the reciprocal group velocity in this band is $k_q' \equiv (d/d\omega)[n(\omega)\omega/c]_{\omega=q\omega_1}$. Other terms are as in Eqs. (1). Clearly, ionization effects are not included in this model, and physical situations where ionization occurs must be treated differently.

Assuming the same fundamental pulse (2) as for THG, the qth harmonic pulse takes the form

$$u_q(\omega,k_x,k_y,z) = i\frac{(q\omega_1/c)^2}{qk_1k_q}\chi^{\mathrm{qHG}}\frac{2\pi/\sqrt{q}}{t_p^{q-1}}\left(\frac{k_1z_RE_1}{\pi^{3/2}}\right)^{q/2}\exp\left[-\frac{t_p^2\omega^2}{2q}-\frac{z_R(k_x^2+k_y^2)}{2qk_1}+i\left(k_q'\omega-\frac{k_x^2+k_y^2}{2k_q}\right)z\right]$$
$$\times\int_{z_0}^{z}\frac{1}{(z_R+iz')^{q-1}}\exp\left\{i\left[(qk_1-k_q)\left(1+\frac{k_x^2+k_y^2}{2qk_1k_q}\right)+(k_1'-k_q')\omega\right]z'\right\}dz'. \quad (19)$$

In the case of far-field to far-field propagation, the integral is soluble because the integrand is analytic except for a pole of order $(q-1)$ at $z=iz_R$,

$$u_q(\omega,k_x,k_y,z) = i\frac{(q\omega_1/c)^2}{qk_1k_q}\chi^{\mathrm{qHG}}\frac{(2\pi)^2/\sqrt{q}}{(q-2)!}\frac{1}{t_p^{q-1}z_R^{q-2}}\left(\frac{k_1z_RE_1}{\pi^{3/2}}\right)^{q/2}\times\phi^{q-2}\exp\left[-\phi-\frac{t_p^2\omega^2}{2q}-\frac{z_R(k_x^2+k_y^2)}{2qk_1}+i\left(k_q'\omega-\frac{k_x^2+k_y^2}{2k_q}\right)z\right]H(\phi) \quad (20a)$$

with dimensionless mismatch

$$\phi=\phi(\omega,k_x,k_y)\equiv\left[(qk_1-k_q)\left(1+\frac{k_x^2+k_y^2}{2qk_1k_q}\right)+(k_1'-k_q')\omega\right]z_R. \quad (20b)$$

In the case of focus to far-field propagation, the integral may be integrated by parts recursively, until it can be expressed as a special function,

$$u_q(\omega,k_x,k_y,z) = \frac{(q\omega_1/c)^2}{qk_1k_q}\chi^{\mathrm{qHG}}\frac{2\pi/\sqrt{q}}{(q-2)!}\frac{z_R^2}{t_p^{q-1}}\left(\frac{k_1E_1}{z_R\pi^{3/2}}\right)^{q/2}\times\left[\sum_{n=0}^{q-3}(q-3-n)!\,\phi^n-\frac{\mathrm{Ei}(\phi)+i\pi H(\phi)}{\exp(\phi)}\phi^{q-2}\right]\times\exp\left[-\frac{t_p^2\omega^2}{2q}-\frac{z_R(k_x^2+k_y^2)}{2qk_1}+i\left(k_q'\omega-\frac{k_x^2+k_y^2}{2k_q}\right)z\right]. \quad (21)$$

The elaboration of the general higher harmonic generation case to all the various limits is straightforward and lengthy, so we do not give this explicitly.

VI. CONCLUSIONS

We showed that in third-harmonic generation by focused pulses, group-velocity mismatch, as well as the familiar phase-velocity mismatch, can strongly affect the third-harmonic pulse produced. We limited the analysis to cases in which dispersion (within the envelope of the fundamental and third harmonic) and Kerr effects may be neglected. Whether dispersion and Kerr effects are small enough, compared to effects of phase- and group-velocity mismatch and of diffraction, will depend on the medium and the duration and intensity of the pulse.

There is a characteristic time scale determined by the difference between the group velocities at the fundamental and its third harmonic, and the Rayleigh range. Pulses with temporal width much greater than the characteristic time scale behave like continuous-wave (monochromatic) beams; pulses with temporal width in the range of, or shorter than, the characteristic time scale exhibit qualitatively different behavior. We detailed the behavior qualitatively and quantitatively—giving analytic solutions—for pulses in which most of the energy is in the fundamental beam.

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