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Group velocity difference effect on incoherent sum-frequency generation spectrum

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The spectral distribution of optical sum-frequency generation (SFG) by incoherent non-linear mixing of two chaotic beams with identical spectral width η is calculated theoretically in the first approximation of the iterative method including dispersion of the non-linear medium and the difference κ between the group velocity of the input beams. It is shown that for the same group velocities ($\kappa = 0$) of the input beams in non-linear medium the resultant field has a spectral width narrower than η, and if a phase mismatch is present the spectral maximum of the resultant field is shifted towards lower or higher frequencies according to the sign of the phase mismatch. If the input beams have different group velocities ($\kappa \neq 0$) in the non-linear medium, then the spectral distribution of SFG is strongly dependent on η. For small η the spectral distribution of SFG is the same irrespective of κ. For large η the difference in group velocities broadens the spectrum of SFG and for very large differences in group velocity the SFG has a spectral width identical to that of the input beams.

1. Introduction

Optical wave interaction in a non-linear medium leads to wave mixing which is a non-linear phenomenon giving rise to generation of waves at sum and difference frequencies. The process of optical sum-frequency generation (SFG) is one of the most useful non-linear optical effects, for example in extending the tunable range of lasers to shorter wavelengths. The physical interpretation of sum-frequency generation is straightforward. The laser photons at the lower frequencies ω_1 and ω_2, by way of non-linear optical interaction in a quadratic medium, generate photons at the higher sum-frequency ω_3 ($\omega_3 = \omega_1 + \omega_2$).

The basic classical theory of optical sum-frequency generation is due to Armstrong *et al.* [1] and has been verified in experiments by Bass *et al.* [2]. In frequency-conversion experiments one usually deals with single or multimode laser radiations with fluctuating phases and amplitudes. Therefore, it is important to understand the influence of the light fluctuations on non-linear optical processes. The influence of partial coherence of the generating radiations was first discussed by Ducuing and Bloembergen [3] and Akhmanov and Chirkin

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[4] in second-harmonic generation (degenerate sum-frequency generation) and by Chmela [5] in parametric up-conversion. Various aspects of incoherent three-wave mixing and second-harmonic generation with phase fluctuations were studied in [6–8]. In particular, as was shown in [7], the efficiency of three-wave mixing in non-linear dispersive medium with one coherent beam and one spectrally narrow chaotic input beam increases with increasing difference between the group velocities of the input beams, but decreases for large spectral width of the chaotic beam. Much attention has recently been paid to the incoherent parametric non-linear optical interaction of short laser pulses in connection with the limitation imposed on the conversion efficiency by group velocity mismatch [9–11].

The statistical behaviour of light in incoherent non-linear optical interactions of radiations possessing well-defined spectral widths in dispersive medium represents a peculiar topic among other statistical phenomena in non-linear optical processes [4, 12–15]. In particular, the theoretical description of incoherent non-linear optical interactions, including partial coherence of interacting radiations and medium dispersion, is mathematically rather more exacting than for coherent interactions.

In this paper we study the spectral distribution of sum-frequency generation with chaotic input radiations when the generating beams have arbitrary intensities and finite spectral widths. Moreover, differences between the group velocities of the input beams in dispersive medium and phase mismatch are assumed. We start from the set of first-order differential equations for complex, slowly varying field amplitudes describing the incoherent quadratic non-linear optical interaction. The solution for the spectral distribution of SFG is derived to a first approximation by the iterative method [4].

2. Spectral distribution of SFG

The aim of this paper is to calculate the spectral distribution of the resultant sum-frequency radiation in non-linear optical interaction of three quasi-monochromatic waves:

$$\boldsymbol{E}_j(\boldsymbol{r}, t) = \boldsymbol{e}_j A_j(\boldsymbol{r}, t)\exp[\mathrm{i}(\boldsymbol{k}_j \cdot \boldsymbol{r} - \omega_j t)] \qquad j = 1, 2, 3 \tag{1}$$

propagating in a dispersive medium, where $\boldsymbol{e}_j$ are the unit polarization vectors and the frequencies ω_1, ω_2 and ω_3 satisfying the resonant frequency condition $\omega_1 + \omega_2 = \omega_3$. The complex, slowly varying field amplitudes $A_j(\boldsymbol{r},t)$ in a dispersive non-linear quadratic medium can be described, from the point of view of classical theory, by means of three coupled first-order partial differential equations [4, 5]:

$$\begin{aligned}
\operatorname{grad} A_1 \cdot \boldsymbol{f}_1 + u_1^{-1}\,\partial A_1/\partial t &= \mathrm{i}\alpha_1 A_3 A_2^* \exp(\mathrm{i}\,\Delta k\, z) \\
\operatorname{grad} A_2 \cdot \boldsymbol{f}_2 + u_2^{-1}\,\partial A_2/\partial t &= \mathrm{i}\alpha_2 A_3 A_1^* \exp(\mathrm{i}\,\Delta k\, z) \\
\operatorname{grad} A_3 \cdot \boldsymbol{f}_3 + u_3^{-1}\,\partial A_3/\partial t &= \mathrm{i}\alpha_3 A_1 A_2 \exp(-\mathrm{i}\,\Delta k\, z)
\end{aligned} \tag{2}$$

where u_j ($j = 1, 2, 3$) are the group velocities in the ray directions $\boldsymbol{f}_j$ of the individual waves, α_j are the coupling constants which depend on the second-order susceptibility of the non-linear medium [5], and $\Delta k = (k_3 - k_2 - k_1)_z$ represents the normal component of the wave-mismatch vector. The co-ordinates of the system (x, y, z) are oriented so that the z-axis is normal to the first boundary of the non-linear medium.

The homogeneous Equations 2 for non-perturbed amplitudes have the well-known solutions

$$A_{j,0}(\boldsymbol{r}, t) = A_{j,0}\left(t - \frac{\boldsymbol{f}_j \cdot \boldsymbol{r}}{u_j}\right) \qquad j = 1, 2, 3 \tag{3}$$

Thus, when considering the interaction of spatially unlimited plane waves, we can write the boundary conditions for SFG in the form

$$A_{j,0}(\boldsymbol{r}, t) = A_{j,0}\left(t - \frac{\boldsymbol{f}_j \cdot \boldsymbol{r}}{u_j}\right) \quad \text{for} \quad z \leqslant 0 \; (j = 1, 2) \tag{4}$$

and $A_{3,0}(\boldsymbol{r}, t) = 0$ for $z \leqslant 0$.

In order to study the spectral distribution of SFG we have to know the two-time second-order correlation function $\langle A_3(\boldsymbol{r}, t) A_3^*(\boldsymbol{r}, t + \tau)\rangle$ for the complex amplitudes of the resultant field. To this aim we shall solve the system of three coupled equations (Equations 2) by the iterative method [4]. We restrict our solution to the first step of the iterative method. In this approximation we assume that neither of the generating beams at ω_1 and ω_2 are perturbed by the non-linear interaction, and that they are given by Equation 3. For the resultant field the first-step iterative solutions of Equations 2 with the boundary conditions 4 leads to the following expression for the stochastic amplitude:

$$A_3(\boldsymbol{r}, t) = \mathrm{i}\sigma_3 \int_0^z \mathrm{d}\mu \, \mathrm{e}^{-\mathrm{i}\Delta k \mu} A_{1,0}\left(t - \frac{\boldsymbol{f}_1 \cdot \boldsymbol{r}}{u_1} - \varepsilon_{13} z + \varepsilon_{13}\mu\right) A_{2,0}\left(t - \frac{\boldsymbol{f}_2 \cdot \boldsymbol{r}}{u_2} - \varepsilon_{23} z + \varepsilon_{23}\mu\right) \tag{5}$$

where $\sigma_3 = \alpha_3/\cos\beta_3$, with β_3 the refractive angle in the ray direction $\boldsymbol{f}_3$, and $\varepsilon_{j3} (j \neq 3)$ are typical dispersion coefficients dependent on the group velocity difference between the generating and resultant fields, and are defined as

$$\varepsilon_{j3} = \frac{1}{\cos\beta_3}\left(\frac{1}{u_3} - \frac{\cos\varrho_{j3}}{u_j}\right) \qquad j = 1, 2 \tag{6}$$

where ϱ_{j3} are the divergence angles between the two ray directions $\boldsymbol{f}_j$ and $\boldsymbol{f}_3$. Hence, from Equation 5 and assuming that the two input beams at ω_1 and ω_2 are initially not correlated, we find that the two-time second-order correlation function of the complex amplitudes of SFG has the form

$$\begin{aligned}\langle A_3(\boldsymbol{r}, t) A_3^*(\boldsymbol{r}, t + \tau)\rangle = {} & \sigma_3^2 \int_0^z \mathrm{d}\mu_1 \int_0^z \mathrm{d}\mu_2 \exp[-\mathrm{i}\Delta k(\mu_1 - \mu_2)] \\ & \times \langle A_{1,0}(t) A_{1,0}^*[t + \tau + \varepsilon_{13}(\mu_1 - \mu_2)]\rangle \\ & \times \langle A_{2,0}(t) A_{2,0}^*[t + \tau + \varepsilon_{23}(\mu_1 - \mu_2)]\rangle\end{aligned} \tag{7}$$

From this equation it is evident that, for explicit calculations of the spectral distribution of SFG generated in an incoherent non-linear optical interaction, it is necessary to know the mean values $\langle A_{j,0}(t_1) A_{j,0}^*(t_2)\rangle$. To this aim, we assume that both input beams are chaotic with identical finite spectral half-width Γ and obey the factorization relationship [15]

$$\langle A_{j,0}(t_1) A_{j,0}^*(t_2)\rangle = \langle J_{j,0}\rangle \exp[-\Gamma(t_2 - t_1)] \tag{8}$$

with $\langle J_{j,0}\rangle = \langle A_{j,0} A_{j,0}^*\rangle$ describing the initial intensity of the input beams.

On deriving the Fourier transform of Equation 7 with Equation 8 and assuming that ε_{13} and ε_{23} are positive and that $\varepsilon_{13} > \varepsilon_{23}$, we obtain the following formula for the spectral distribution of SFG:

$$\Phi(\Omega) = \Phi_0 \left(\frac{4\eta^2\kappa^3\varphi}{[(\eta\kappa)^2 + (\Omega + \Delta)^2][(\eta\kappa)^2 + (\Omega + \Delta - \kappa\Omega)^2]} + \frac{8\eta^2 A e^{-\eta\kappa\varphi}\sin(\Omega + \Delta)\varphi - 4\eta B[1 - e^{-\eta\kappa\varphi}\cos(\Omega + \Delta)\varphi]}{\Omega[4\eta^2 + \Omega^2][(\eta\kappa)^2 + (\Omega + \Delta)^2]^2} - \frac{8\eta^2 A' e^{-\eta\kappa\varphi}\sin(\Omega + \Delta - \kappa\Omega)\varphi + 4\eta B'[1 - e^{-\eta\kappa\varphi}\cos(\Omega + \Delta - \kappa\Omega)\varphi]}{\Omega[4\eta^2 + \Omega^2][(\eta\kappa)^2 + (\Omega + \Delta - \kappa\Omega)^2]^2} \right) \tag{9}$$

with

$$\begin{aligned} \Phi_0 &= \frac{\varepsilon_{13}}{2\pi\langle J_{1,0}\rangle^{1/4}\langle J_{2,0}\rangle^{1/4}\sigma_3} \\ A &= (\eta\kappa)^2 - (\Omega + \Delta)(\Omega + \Delta + \kappa\Omega) \\ A' &= A + 2\kappa\Omega(2\Omega + 2\Delta - \kappa\Omega) \\ B &= \Omega\eta^2\kappa^2 + (\Omega + \Delta)[4\eta^2\kappa - \Omega(\Omega + \Delta)] \\ B' &= B - \kappa(4\eta^2 - \Omega^2)(2\Omega + 2\Delta - \kappa\Omega) \end{aligned}$$

where, for simplicity, we have introduced the notation

$$\begin{aligned} \varphi &= \langle J_{1,0}\rangle^{1/4}\langle J_{2,0}\rangle^{1/4}\sigma_3 z \qquad & \Delta &= \frac{\Delta k}{\langle J_{1,0}\rangle^{1/4}\langle J_{2,0}\rangle^{1/4}\sigma_3} \\ \eta &= \frac{\Gamma|\varepsilon_{13}|}{\langle J_{1,0}\rangle^{1/4}\langle J_{2,0}\rangle^{1/4}\sigma_3} \qquad & \Omega &= \frac{\varepsilon_{13}(\omega - \omega_3)}{\langle J_{1,0}\rangle^{1/4}\langle J_{2,0}\rangle^{1/4}\sigma_3} \\ & & \kappa &= \frac{\varepsilon_{13} - \varepsilon_{23}}{\varepsilon_{13}} \end{aligned} \tag{10}$$

The explicit iterative solution (Equations 9) shows the resonance structure of the SFG spectrum which is dependent on four parameters: η, Δ, κ and φ. The parameter η describes the spectral half-width Γ of the input beams, whereas Δ describes the normalized wave-mismatch vector. The parameter κ is dependent on the difference between the group velocities of the input waves; $\kappa = 0$ for identical group velocities of input waves, whereas $\kappa = 1$ for a very large difference between them. The solution (Equations 9) is valid for all values of the input intensities $\langle J_{1,0}\rangle$ and $\langle J_{2,0}\rangle$, but is of limited applicability with respect to the parameter φ, which describes the normalized thickness of the non-linear medium.

The solution (Equations 9) shows for $\kappa \neq 0$ a Lorentzian structure of the SFG spectrum with peaks at $\Omega_1 = 0$, $\Omega_2 = -\Delta$ and $\Omega_3 = -\Delta/(1 - \kappa)$. The peaks at Ω_2 and Ω_3 have a spectral width dependent on κ. Since $\kappa \in \langle 0, 1\rangle$, these peaks are narrower than the width of the input beams, and are the same only for very large differences between the group velocities ($\kappa \approx 1$). If the group velocities of the input beams are identical ($\kappa = 0$) in the non-linear medium, the solution reduces to

$$\Phi(\Omega) = \Phi_0 \left[\frac{4\eta\varphi^2}{(4\eta^2 + \Omega^2)} \left(\frac{\sin\frac{1}{2}(\Omega + \Delta)\varphi}{\frac{1}{2}(\Omega + \Delta)\varphi} \right)^2 \right] \tag{11}$$

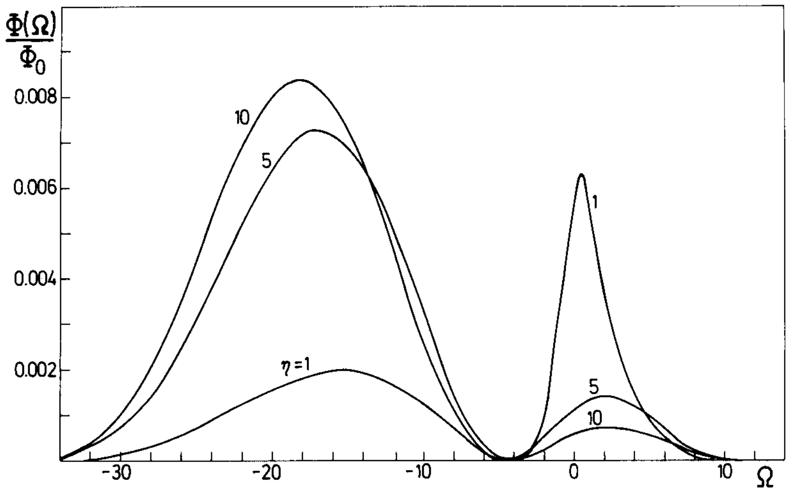

Figure 1 Spectral distribution of SFG for $\Delta = 20$, $\varphi = 0.4$, $\kappa = 0$ and different values of the spectral width η.

If follows from Equation 11 that the central maximum of SFG becomes narrow compared with the spectral distribution of the input beams, and its location and width are dependent on the function $\sin^2 x/x^2$, which has a strong peak at $x = 0$. For $\Delta = 0$ this maximum is located at $\Omega = 0$, but for $\Delta \neq 0$ the spectrum is asymmetric and the main peak is located at $\Omega = -\Delta$ according to the sign of Δ. For sign $\Delta = +1$, the central maximum is shifted towards lower frequencies, whereas the central maximum is shifted towards higher frequencies if sign $\Delta = -1$. This effect is similar to that discussed in [5] for parametric up-conversion. The intensity of this shifted maximum is dependent on the spectral width of the input beams. This is shown in Fig. 1, where the spectral distribution of Equation 11 is plotted for $\Delta = 20$, $\varphi = 0.4$ and different values of η. For small spectral width η of the input beams the spectrum consists of one high peak at $\Omega = 0$ and a small side peak at $\Omega = -\Delta$. As η increases, the intensity of the side peak increases whereas the peak at $\Omega = 0$ decreases. The dependence of SFG on the spectral width of the input beams can be explained within the framework of the phase mismatch effect. At the beginning of the process the phase mismatch does not markedly affect the non-linear process and the frequency maximum of SFG appears at $\omega_{3,0} = \omega_{1,0} + \omega_{2,0}$, where $\omega_{j,0}$ are mean values of the frequencies ω_j. For considerable large phase mismatch ($|\Delta| \gg 1$), in the further course of SFG only those frequencies will be systematically amplified which satisfy the approximate phase matching condition

$$(\boldsymbol{k}_{\omega_3} - \boldsymbol{k}_{\omega_2} - \boldsymbol{k}_{\omega_1})_z \lesssim \pi \qquad (12)$$

The remaining frequencies which do not satisfy this condition are damped due to the phase mismatch effect, which is a consequence of destructive interference of radiations that are generated at different distances from the non-linear medium boundary. If the spectral width of the input beams is small ($\eta \ll 1$), at the beginning of the SFG process practically all of the energy is converted into the resultant field at $\Omega = 0$, and only a small part of the energy remains for the further course of SFG and is converted with the condition of Equation 12 giving a small peak at $\Omega = -\Delta$ (see Fig. 1). Conversely, for large η, at the beginning of the SFG process a relatively small part of the input energy is converted into SFG and the

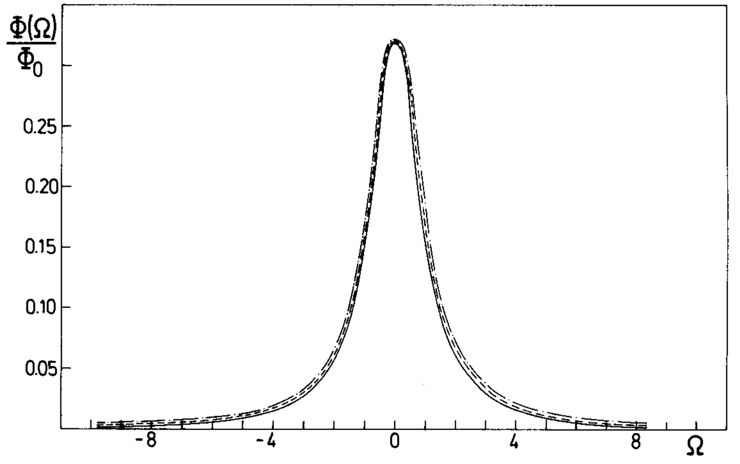

Figure 2 Spectral distribution of SFG for $\Delta = 0$, $\varphi = 0.4$, $\eta = 0.5$ and different group velocities of the input beams: (——) $\kappa = 0$, (- - -) $\kappa = 0.5$ and (—·—) $\kappa = 1$.

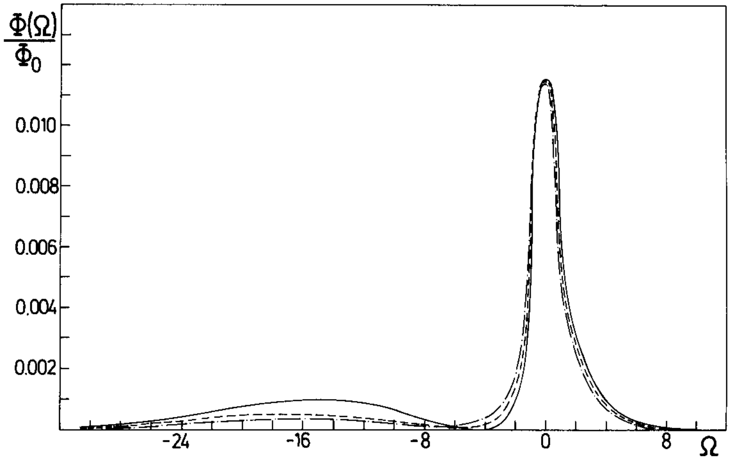

Figure 3 The same as Fig. 2, but for greater phase mismatch ($\Delta = 20$).

remaining, considerably great part of the energy is later converted with approximate phase matching (Equation 12). As η increases, the greater part of the input energy is converted into the resultant beam, leading to the increase of the peak at $\Omega = -\Delta$.

The effect of group velocity difference between the input beams on the spectrum of SFG given by the general solution (Equations 9) is shown in Figs 2 to 5, where $\Phi(\Omega)$ is plotted for $\phi = 0.4$ and different η, Δ and κ. It is obvious from Figs 2 and 3 that for small spectral width η of the input beams, irrespective of Δ, the group velocity difference does not affect the spectrum of SFG. However, for large η the group velocity difference broadens the resultant field considerably, as shown in Figs 4 and 5. This can be explained as follows. For small spectral widths η of the input beams the coherence times or lengths of all of the interacting beams are relatively large, which explains why the behaviour of SFG spectra is very weakly sensitive to the group velocity difference κ. On the other hand, if the spectral

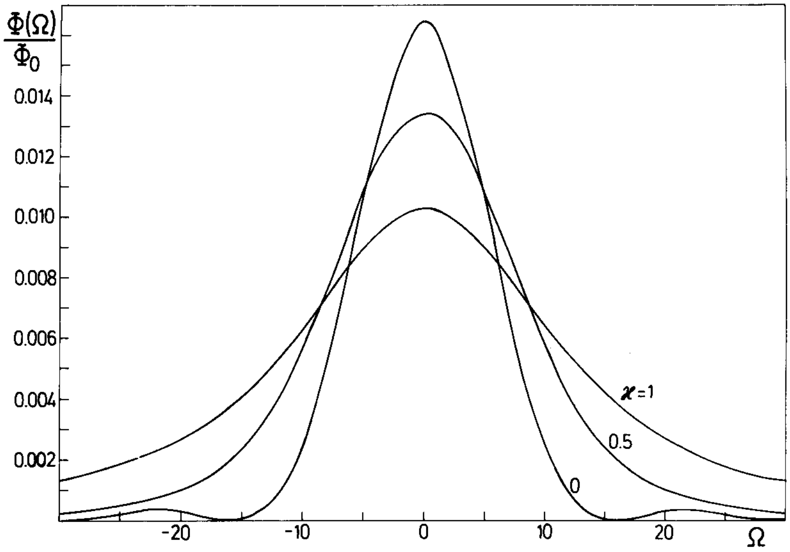

Figure 4 Spectral distribution of SFG for $\Delta = 0$, $\varphi = 0.4$, larger spectral width η ($\eta = 10$) and different group velocities of the input beams.

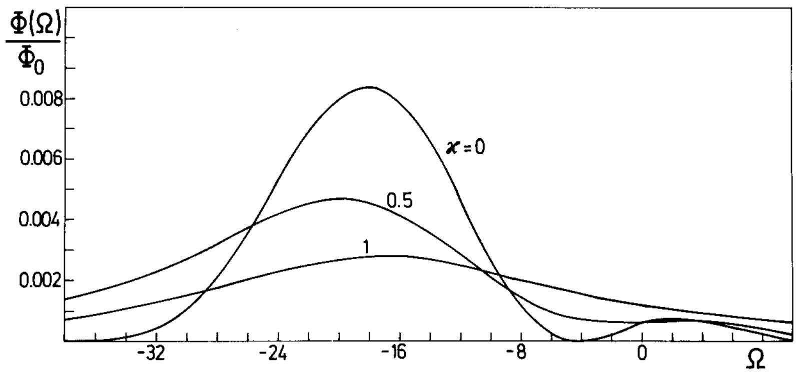

Figure 5 The same as Fig. 4, but for greater phase mismatch ($\Delta = 20$).

widths of the input beams are large ($\eta \gg 1$), then the interacting beams possess relatively very short coherence times or lengths. Consequently, the sub-frequency beams generated, which are considerably time- or spatial-shifted due to the group velocity difference, are mutually incoherent. Therefore, the effects of phase matching or phase mismatch connected with the interference phenomena cannot assert themselves. This explains the generation of relatively broadened sum-frequency spectra for large η and κ, as is seen in Figs 4 and 5.

To conclude, we would point out that sometimes it is useful to calculate the degree of coherence [5, 15] instead of the spectral distribution of the resultant field. For SFG, the degree of coherence can be defined as

$$\gamma_3(\boldsymbol{r}, \tau) = \langle A_3(\boldsymbol{r}, t) A_3^*(\boldsymbol{r}, t + \tau) \rangle / \langle A_3(\boldsymbol{r}, t) A_3^*(\boldsymbol{r}, t) \rangle$$

Obviously, on dividing the spectral distribution (Equations 9) by the intensity of the resultant field $\langle A_3(\boldsymbol{r}, t) A_3^*(\boldsymbol{r}, t)\rangle$, we obtain the spectral distribution of the degree of coherence. In the same way we can define the degree of coherence of the input beams. For chaotic input beams the degree of coherence of the individual beams is simply given by the function $\gamma_{1,2}(\tau) = \exp(-\Gamma\tau)$. The maximum possible spectral width of the sum-frequency radiation generated by incoherent non-linear optical mixing of two chaotic beams is 2Γ. Therefore, we can refer our discussion on the degree of coherence of the generated sum-frequency radiation $\gamma_3(\boldsymbol{r}, \tau)$ to the degree of coherence of the individual input beams. From our solution we find that for $\kappa = 0$ the resultant field in the process of incoherent SFG is more coherent than the input fields, and for small η the group velocity difference between the input beams does not affect this high degree of coherence. However, for large η the group velocity difference lowers the degree of coherence. For very large differences between the group velocities, the degree of coherence of the resultant field is the same as that of the input fields.

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