# ENHANCED INCOHERENT SUM-FREQUENCY GENERATION BY GROUP VELOCITY DIFFERENCE ☆

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We propose a theoretical treatment of optical sum-frequency generation (SFG) by incoherent nonlinear mixing of two beams, the one coherent and the other chaotic. The efficiency of SFG is calculated in the second approximation of the iterative method solving the equations for parametric interaction of the three waves. It is shown that in the case of perfect phase matching the efficiency of SFG increases with increasing spectral width $\eta$ of the input chaotic radiation and increasing difference $\kappa$ between the group velocities of the sub-frequency modes. For small spectral widths of the chaotic sub-frequency radiation the efficiency of SFG is greater than for coherent interaction provided that $\kappa$ is large. For large spectral widths, however, the efficiency of SFG is smaller than for coherent interaction irrespective of $\kappa$.

## 1. Introduction

The process of optical sum-frequency generation (SFG) is a nonlinear phenomenon of great practical importance [1–3], and moreover presents highly interesting theoretical aspects [4]. The process consists in the generation of photons at the higher sum-frequency $\omega_3$ by fusing the photons at the lower frequencies $\omega_1$ and $\omega_2$ ($\omega_3=\omega_1+\omega_2$) by way of nonlinear optical interaction in a quadratic medium.

The classical description of optical sum-frequency generation has been first given by Armstrong et al. [5] and observed experimentally by Bass et al. [6]. The quantum description of this process was given by Agrawal and Mehta [7] and Peřina [8]. The influence of the statistical and coherence properties of the generating radiations was first discussed by Ducuing and Bloembergen [9] and Akhmanov and Chirkin [10] in second-harmonic generation (degenerate sum-frequency generation) and by Chmela [11] in non-degenerate SFG. In frequency-conversion experiments one usually deals with single or multimode laser radiations, while the amplitudes can exhibit substantial light fluctuations. Therefore, it is important to understand the influence of the light fluctuations on nonlinear optical processes. The effects of phase and amplitude fluctuations in the generating fundamental radiations on second-harmonic generation were studied in refs. [12–14]. Eckardt and Reintjes [15] studied second-harmonic generation with phase modulation of the fundamental pulse. Chmela [11] considered SFG by incoherent nonlinear optical fusing of strong coherent radiation and another weak chaotic input beam with a finite spectral width. He has shown that the efficiency of SFG decreases with increasing spectral width of the input radiation and/or increasing dispersion of the medium and is independent of the difference between the group velocities of the interacting radiations.

In this paper we study the influence of field amplitude fluctuations on the SFG process when the input radiations have arbitrary intensities. As we shall see later, in this case the efficiency of SFG is dependent on the difference $\kappa$ between the group velocities of the input radiations and increases with increasing spectral width $\eta$, attains its maximum value for moderate $\eta$, and then decreases for large $\eta$. We start from the set of first-order

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differential equations for complex slowly varying field amplitudes describing the incoherent quadratic nonlinear optical interaction. The solution is found by the iterative method in the second approximation and perfect phase matching is assumed.

It follows from this solution that the mean photon flux (intensity) of output sum-frequency radiation is dependent on the spectral width of the input radiation, the dispersion of the nonlinear medium, and the difference between the group velocities of the input radiations.

## 2. Sum-frequency generation by incoherent interactions

Within the framework of our classical model we consider nonlinear optical interaction of three quasimonochromatic waves propagating in a dispersive medium:

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

where $\boldsymbol{e}_j$ are the unit polarization vectors and $A_j(\boldsymbol{r},t)$ the slowly varying field amplitudes. The frequencies $\omega_1$, $\omega_2$ and $\omega_3$ satisfy the resonant frequency condition

$$\omega_1+\omega_2=\omega_3. \tag{2}$$

The complex amplitudes $A_j(\boldsymbol{r},t)$, $(j=1,2,3)$ in the linear dispersion approximation obey the following equations [10,11]:

$$\operatorname{grad} A_1\cdot\boldsymbol{f}_1+u_1^{-1}\,\partial A_1/\partial t=\mathrm{i}\gamma_1 A_3A_2^*\exp(\mathrm{i}\Delta kz),$$

$$\operatorname{grad} A_2\cdot\boldsymbol{f}_2+u_2^{-1}\,\partial A_2/\partial t=\mathrm{i}\gamma_2 A_3A_1^*\exp(\mathrm{i}\Delta kz),$$

$$\operatorname{grad} A_3\cdot\boldsymbol{f}_3+u_3^{-1}\,\partial A_3/\partial t=\mathrm{i}\gamma_3 A_1A_2\exp(-\mathrm{i}\Delta kz), \tag{3}$$

where $\Delta k=k_3-k_2-k_1$ is the wave vector mismatching, $\gamma_j$ are the coupling constants which depend on the second-order susceptibility of the nonlinear medium [11], and $u_j$ are the group velocities in ray directions $\boldsymbol{f}_j$ of the individual waves. It is obvious from eqs. (3) that, in general, the nonlinear optical interaction in a dispersive medium cannot be simply described in terms of one time $(t)$ or one spatial $(z)$ variable only, as usually done when studying nonlinear optical interactions of monochromatic waves [5]. In general, the field amplitudes $A_j(\boldsymbol{r},t)$ are functions of one temporal variable $t$ and three spatial variables $x$, $y$, $z$. However, the mean values of the intensities $\langle I_j\rangle$ or photon numbers $\langle N_j\rangle$, as well as the higher-order moments of the type $\langle I_j^m I_k^p\rangle$ or $\langle N_j^m N_k^p\rangle$ can be expressed in terms of one temporal variable $t$ or one spatial variable $z$ only, according to the model of interaction considered.

Our aim is to calculate the mean photon flux $\langle N_3(z)\rangle$ instead of the light intensities in the output beam defined as

$$\langle N_3(z)\rangle=(\epsilon_0/\mu_0)^{1/2}[(n_3\cos\delta_3\cos\beta_3)/(2\hbar\omega_3)]\langle A_3A_3^*\rangle \tag{4}$$

(in SI units), where $n_3$ is the linear refractive index in an anisotropic medium, $\delta_3$ is the angle of anisotropy (that between the ray direction $\boldsymbol{f}_3$ and the normal direction $\boldsymbol{s}_3$), and $\beta_3$ is the refractive angle of the ray direction $\boldsymbol{f}_3$. It is necessary to stress here that the photon fluxes represent classical quantities and do not involve any quantum aspects of the field.

In order to calculate the mean photon flux $\langle N_3(z)\rangle$ we have to know the factorisation rules for the mean values $\langle A_{j,0}(t)A_{j,0}^*(t+\tau)\rangle$ and $\langle A_{j,0}(t)A_{j,0}^*(t+\tau)A_{j,0}(t+\tau')A_{j,0}^*(t+\tau'')\rangle$, $(j=1,2)$, where the $A_{j,0}(t)$ are nonperturbed amplitudes for the first step of the iterative solution. In the following, we assume that the input radiation at frequency $\omega_1$ is coherent. We thus have [16]

$$\langle A_{1,0}(t_1)\, A^*_{1,0}(t_2)\rangle = \langle N_1(0)\rangle, \qquad \langle A_{1,0}(t_1)\, A^*_{1,0}(t_2)\, A_{1,0}(t_3)\, A^*_{1,0}(t_4)\rangle = \langle N_1(0)\rangle^2, \tag{5}$$

and the input radiation at frequency $\omega_2$ is chaotic with the finite spectral half-width $\Gamma_2$ introduced as [11,16]

$$\langle A_{2,0}(t_1)\, A^*_{2,0}(t_2)\rangle = \langle N_2(0)\rangle \exp[-\Gamma_2 |t_2 - t_1|],$$

$$\langle A_{2,0}(t_1)\, A^*_{2,0}(t_2)\, A_{2,0}(t_3)\, A^*_{2,0}(t_4)\rangle$$

$$= \langle N_2(0)\rangle^2 \{\exp[-\Gamma_2(|t_2-t_1| + |t_4-t_3|)] + \exp[-\Gamma_2(|t_2-t_3| + |t_4-t_1|)]\}. \tag{6}$$

Both input radiations at frequencies $\omega_1$ and $\omega_2$ are considered as initially not correlated.

Using eqs. (3)–(6) with the boundary conditions for the SFG:

$$A_{j,0}(\boldsymbol{r}, t) = A_{j,0}(t - \boldsymbol{f}_j\cdot\boldsymbol{r}/u_j) \quad \text{for } z \leqslant 0,\ (j=1,2); \qquad A_{3,0}(\boldsymbol{r}, t) = 0 \quad \text{for } z \leqslant 0, \tag{7}$$

after straightforward but lengthy algebraic manipulations and assuming $\Delta k = 0$, we get the following second-approximation iterative solution for the mean photon flux of the sum-frequency wave:

$$\langle N_3(\zeta)\rangle = \langle N_1(0)\rangle^{1/2} \langle N_2(0)\rangle^{1/2} I(\eta,\zeta) - \langle N_1(0)\rangle E_1(\eta,\zeta) - \langle N_2(0)\rangle [E_2(\eta,\zeta) + F_0(\eta,\kappa,\zeta)], \tag{8}$$

with

$$I(\eta,\zeta) = (2/\eta)\zeta - (2/\eta^2)[1 - \exp(-\eta\zeta)], \tag{9a}$$

$$E_1(\eta,\zeta) = (2/3\eta)\zeta^3 - (4\zeta/\eta^3)[1 + \exp(-\eta\zeta)] + (8/\eta^4)[1 - \exp(-\eta\zeta)], \tag{9b}$$

$$E_2(\eta,\zeta) = (2/\eta^2)\zeta^2 - (6/\eta)\zeta + (1/\eta^4)[7 + \exp(-2\eta\zeta) - 8\exp(-\eta\zeta)], \tag{9c}$$

$$F_0(\eta,\kappa,\zeta) = \frac{2}{\kappa\eta^2}\zeta^2 - \frac{2(\kappa^2+2)}{\kappa^2\eta^3}\zeta - \frac{4(1-\kappa)\zeta}{(1-2\kappa)\eta^3}\exp(-\eta\zeta) + \frac{4\kappa^3 - \kappa^2 + 4}{\kappa^3\eta^4}$$

$$+ \frac{\exp(-2\eta\kappa\zeta)}{\kappa(1-2\kappa)^2\eta^4} - \frac{4}{\kappa^3\eta^4}\exp(-\kappa\eta\zeta) - \frac{4(4\kappa^2 - 5\kappa + 2)}{\eta^4(1-2\kappa)^2}\exp(-\eta\zeta), \tag{9d}$$

In eqs. (8)–(9) we have introduced the notation

$$\zeta = \langle N_1(0)\rangle^{1/4} \langle N_2(0)\rangle^{1/4} \mu z, \qquad \eta = \Gamma_2 |\epsilon_{23}| / \langle N_1(0)\rangle^{1/4} \langle N_2(0)\rangle^{1/4} \mu, \qquad \kappa = \epsilon_{12}/|\epsilon_{23}|, \tag{10}$$

where $\mu$ denotes the nonlinear coupling constant [11] and the $\epsilon_{ij}$ are coefficients dependent on the group velocity differences and defined as

$$\epsilon_{ij} = (1/\cos\beta_j)(1/u_j - \cos\alpha_{ij}/u_i), \qquad (i \neq j) \tag{11}$$

where $\alpha_{ij}$ are the divergence angles between the two ray directions $\boldsymbol{f}_i$ and $\boldsymbol{f}_j$.

The iterative solution (8) is dependent on four parameters, namely, $\eta$, $\zeta$, $\epsilon_{23}$, and $\kappa$. The parameter $\eta$ describes the spectral half-width $\Gamma_2$ of the input radiation at $\omega_2$ whereas $\epsilon_{23}$ describes the difference between the group velocities of the sub-frequency wave at $\omega_2$ and the sum-frequency wave at $\omega_3$. The parameter $\kappa$ is dependent on the difference between the group velocities of the input waves. The solution (8) is valid for all values of $\langle N_1(0)\rangle$ and $\langle N_2(0)\rangle$, but is of limited applicability with respect to the parameter $\zeta$.

For coherent interaction (for $\eta = 0$) the solution (8) is independent of $\epsilon_{23}$ and $\kappa$, and has the simple form:

$$\langle N_3(\zeta)\rangle = \langle N_1(0)\rangle^{1/2} \langle N_2(0)\rangle^{1/2}\zeta^2 - (\langle N_1(0)\rangle + 2\langle N_2(0)\rangle)\,\tfrac{1}{3}\zeta^4. \tag{12}$$

In this case the efficiency of SFG depends on the normalized thickness $\zeta$ of the nonlinear medium only.

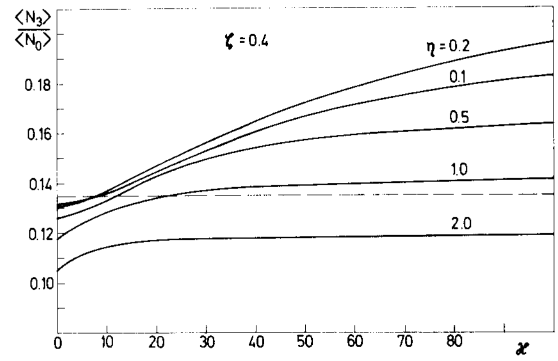


Fig. 1. Relative mean photon flux $\langle N_3(\zeta)\rangle/\langle N_0\rangle$, $(\langle N_0\rangle=\langle N_1(0)\rangle=\langle N_2(0)\rangle)$ in the sum-frequency radiation as a function of $\kappa$ for normalized thickness $\zeta=0.4$ of the nonlinear medium and different normalized spectral widths $\eta$. Dashed line marks the value of the relative mean photon flux for coherent interaction.

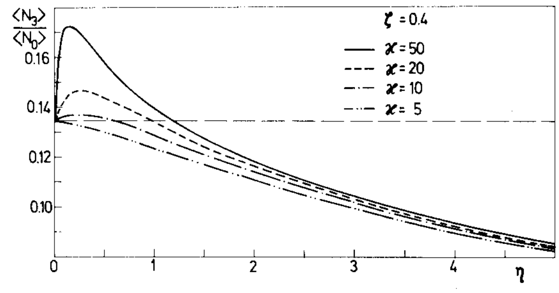


Fig. 2. Relative mean photon $\langle N_3(\zeta)\rangle/\langle N_0\rangle$ versus normalized spectral width $\eta$ for $\zeta=0.4$ and different $\kappa$. Dashed line marks the value of the relative mean photon flux for coherent interaction.

The total mean photon flux $\langle N_3(\zeta)\rangle$ of the sum-frequency radiation, given by eq. (8), is plotted in fig. 1 for $\zeta=0.4$, several values of $\eta$ and different $\kappa$. It is obvious that for small $\eta$ the efficiency of SFG is enhanced compared with coherent interaction. As $\eta$ increases, the SFG efficiency increases, reaching its maximum value, and then decreases for larger $\eta$. This is shown in fig. 2, where $\langle N_3(\zeta)\rangle$ is plotted versus $\eta$ for $\zeta=0.4$ and different values of $\kappa$. The maximum value $\eta_0$ for which the SFG efficiency is enhanced with respect to coherent interaction is dependent on $\kappa$ (fig. 2) and the normalized thickness $\zeta$ of the nonlinear medium. This is shown in fig. 3, where the limiting value $\eta_0$ of the spectral width of the input radiation at frequency $\omega_2$ for which the effect of enhancement of the SFG efficiency appears, is plotted versus $\zeta$. As $\zeta$ increases, the limiting value $\eta_0$ increases linearly.

The enhancement of SFG efficiency with increasing $\kappa$ for small values $\eta$ $(\eta<\eta_0)$ can be explained within the framework of light statistics. As it has been shown in ref. [17], during the SFG process for one coherent and one chaotic input radiation with the same group velocities $(\kappa=0)$ in the nonlinear medium, anticorrelation is generated between the input radiations. This effect leads to a decrease of efficiency of SFG process.

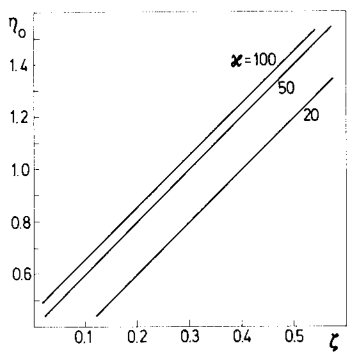


Fig. 3. Limiting value $\eta_0$ of the normalized spectral widths of input chaotic radiation for which the effect of enhanced SFG efficiency appears, versus the normalized thickness $\zeta$ of the nonlinear medium, for different $\kappa$.

If there is a difference between the group velocities ($\kappa \neq 0$) in the nonlinear medium, the generating radiations become time or spatially shifted, so that their natural anticorrelation is compressed or can be completely smoothed out for large $\kappa$ ($\kappa \gg 1$), leading to strong correlation between the input radiations. This strong correlation gives an increase of the SFG efficiency. However, if the spectral width of the chaotic sub-frequency input radiation is considerable ($\eta > \eta_0$), the SFG efficiency decreases with increasing $\eta$ (see fig. 2), irrespective of the value of $\kappa$. This is a consequence of the phase mismatching effect: in fact phase matching can be exactly adjusted for the central maximum frequencies $\omega_{2,0}$ and $\omega_{3,0}$ only ($k_{\omega_{3,0}} - k_{\omega_1} - k_{\omega_{2,0}} = 0$). In this case, since the spectral distribution in the sub-frequency radiation at $\omega_2$ (and consequently also in the sum-frequency radiation at $\omega_3$) is considerably extensive, only relatively small intervals of the frequencies $\omega_2$ and $\omega_3$ satisfy the approximate phase matching condition for the effective SFG: $(k_{\omega_{3,0} \pm \Delta\omega} - k_{\omega_1} - k_{\omega_{2,0} \pm \Delta\omega}) z \ll 1$. The remaining frequencies are phase mismatched, meaning that a considerable amount of the sub-frequency radiation energy is converted into the sum-frequency radiation energy with a variously small efficiency and spatial period according to the frequency difference $\Delta\omega = |\omega_{2,0} - \omega_2|$.

Of course, both above mentioned effects manifest themselves simultaneously. The limiting value $\eta_0$ represents the situation of SFG at which both effects cancel out mutually. For $\eta < \eta_0$, the favourable group velocity dispersion effect predominates over the damping phase mismatching effects, and vice versa for $\eta > \eta_0$.

## 3. Summary

We have studied the problem of sum-frequency generation (SFG) by incoherent nonlinear optical mixing of second-order coherent input radiation and chaotic input radiation of finite spectral width. The efficiency of SFG is described by the mean photon flux $\langle N_3(z) \rangle$ of output radiation and is dependent on the spectral width of the input radiation as well as on the dispersion properties of the nonlinear medium. The iterative method [10] is used to calculate the mean photon flux $\langle N_3(z) \rangle$ in the output sum-frequency radiation.

Our calculations show that the SFG efficiency is strongly dependent on the normalized spectral width $\eta$ of the input chaotic radiation as well as on the difference between the group velocities of the sub-frequency waves characterized by $\kappa$. For small spectral widths $\eta$ of the input radiation the SFG efficiency increases with increasing $\kappa$ (see fig. 1) and next decreases for larger values of $\eta$. For small $\kappa$ or large spectral widths $\eta$ the SFG efficiency is smaller than for coherent interaction, albeit for moderate $\eta$ and large $\kappa$ the efficiency of SFG is greater than for coherent interaction. The limiting value $\eta_0$ (the maximal value of $\eta$ for which the SFG efficiency is enhanced with respect to coherent interaction) depends on the normalized thickness $\zeta$ of the nonlinear medium and increases linearly with increasing $\zeta$.

Two effects influence the efficiency of the SFG process: correlations between the input radiations, and phase mismatching.

At small $\eta$ the former effect predominates for large $\kappa$ leading to an increase of the SFG efficiency. As $\eta$ increases the latter effect becomes predominant leading to a decrease of SFG efficiency. This results from the fact that for large $\eta$ only a small part of the input energies is phase-matched so as to be available for the SFG process.

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