

CLASSICAL THEORY OF NON-DEGENERATE  
SUM-FREQUENCY GENERATION BY INCOHERENT  
NONLINEAR OPTICAL MIXING OF COHERENT  
AND CHAOTIC RADIATIONS

I. GENERAL SOLUTION\*)

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The incoherent nonlinear optical sum-frequency generation with non-monochromatic initially uncorrelated sub-frequency input radiations in a dispersive medium is studied in this paper. The efficiency of the process is calculated in the second approximation, whilst the spectral distribution of generated radiation is merely described in terms of the first approximation of the iterative method. The calculations of the efficiency of nonlinear process and the spectral distribution of sum-frequency radiation are performed for one coherent and the other chaotic input radiations, and for both chaotic input radiations, respectively.

1 INTRODUCTION

The statistical behaviour of light in incoherent nonlinear optical interactions of radiations possessing defined spectral widths in dispersive media (cf. [1-3]) represents a peculiar topic among the class of other statistical phenomena in nonlinear optical processes (for reviews, see refs. [4-14]). Especially, the teoretical description of incoherent nonlinear optical interactions, including spectral distributions of interacting radiations and medium dispersion, is mathematically rather exacting compared with the coherent interactions.

The basic classical theory has been developed by Akhmanov and Chirkin et al. (see [1] and refs. cited therein). The second harmonic generation with chaotic (gaussian) fundamental input radiation was studied by Akhmanov et al. [1, 15] and phase fluctuations were considered by Dutta [16] in the first approximation of the iterative method. Various aspects of incoherent parametric amplification and three wave mixing were treated in [2, 16-21] and stimulated Raman scattering in the field of a noise pump was studied in [22]. Recently, great attention has been paid to the incoherent parametric nonlinear optical interactions of short light pulses in connection with the limitation of conversion efficiency due to the phase mismatch (see e.g. [23-35]).

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The first successful approximate quantum description of the incoherent nonlinear optical interaction was framed by Hong and Mandel [36], who studied the correlation between sub-frequency modes in the parametric down conversion from quantum noise. The theoretically predicted effect [36] was verified by the experimental measurements by Friberg et al. [37].

In this paper we consider the classical evolution of the field in the incoherent non-degenerate nonlinear optical sum-frequency generation with coherent and chaotic sub-frequency input radiations. The chaotic input radiation is supposed to possess a finite spectral width and the dispersion of nonlinear medium is included in our treatment as well. The efficiency of the process is calculated in the second approximation of the iterative method, whilst the spectral distribution of the resulting sum-frequency radiation is described in terms of the first approximation of the iterative solution only. A special case of this problem, namely, the parametric up-conversion of weak chaotic radiation by the incoherent nonlinear optical mixing with strong second-order coherent pumping radiation was considered in [21].

The degenerate case of incoherent second harmonic generation with chaotic fundamental input radiation is not considered here, since it has been sufficiently described by Akhmov and Chirkin [1] in terms of the first-order approximate iterative solution, which also includes the dominant fourth-order statistical behaviour of the field (effects of intensity or photon distribution) at the beginning of the process.

The topic of the article is closely related to the previous papers [38, 39], in which the coherent sum-frequency generation with coherent and chaotic input radiations was studied.

## 2. GENERAL METHOD

The incoherent non-degenerate nonlinear optical interaction of three quasimonochromatic plane waves

$$(I.1) \quad \mathbf{E}_j = \frac{1}{2} \mathbf{e}_j A_j(t, \mathbf{r}) \exp [i(\mathbf{k}_{j,0} \cdot \mathbf{r} - \omega_{j,0}t)] + \text{c.c.} \quad (j = 1, 2, 3),$$

whose mean frequencies satisfy the resonance condition

$$(I.2) \quad \omega_{1,0} + \omega_{2,0} = \omega_{3,0},$$

in a dispersive nonlinear quadratic medium can be described, from the point of view of classical theory, by means of three coupled first-order partial differential equations for complex amplitudes (see e.g. [1, 21, 24, 38, 40])

$$(I.3a) \quad \mathbf{grad} A_1 \cdot \mathbf{f}_1 + \frac{1}{u_{rg1}} \frac{\partial A_1}{\partial t} = i\gamma_1 A_3 A_2^* \exp(i \Delta k z),$$

$$(I.3b) \quad \mathbf{grad} A_2 \cdot \mathbf{f}_2 + \frac{1}{u_{rg2}} \frac{\partial A_2}{\partial t} = i\gamma_2 A_3 A_1^* \exp(i \Delta k z),$$

$$(I.3c) \quad \mathbf{grad} A_3 \cdot \mathbf{f}_3 + \frac{1}{u_{rg3}} \frac{\partial A_3}{\partial t} = i\gamma_3 A_1 A_2 \exp(-i \Delta k z).$$

The following notation has been used:  $\mathbf{f}_j$  are ray directions and  $u_{rgj}$  are group velocities in ray directions of individual waves,  $\gamma_j$  are nonlinear coupling constants given by

$$(I.4) \quad \gamma_j = \left(\frac{\mu_0}{\epsilon_0}\right)^{1/2} \frac{\omega_{j,0} K}{n_j \cos \delta_j} \quad (j = 1, 2, 3),$$

with

$$(I.5) \quad \begin{aligned} K &= \frac{1}{2} \mathbf{e}_1 \cdot \chi^{(2)}(\omega_{1,0} = \omega_{3,0} - \omega_{2,0}): \mathbf{e}_3 \mathbf{e}_2 = \\ &= \frac{1}{2} \mathbf{e}_2 \cdot \chi^{(2)}(\omega_{2,0} = \omega_{3,0} - \omega_{1,0}): \mathbf{e}_3 \mathbf{e}_1 = \\ &= \frac{1}{2} \mathbf{e}_3 \cdot \chi^{(2)}(\omega_{3,0} = \omega_{1,0} + \omega_{2,0}): \mathbf{e}_1 \mathbf{e}_2, \end{aligned}$$

where  $\epsilon_0$  is the electric permittivity and  $\mu_0$  magnetic permeability of vacuum in SI units,  $n_j$  are linear indices of refraction in an anisotropic medium and  $\delta_j$  are angles of anisotropic divergency, i.e. the angles between the ray directions  $\mathbf{f}_j$  and the normal directions  $\mathbf{s}_j$ , respectively,  $\chi^{(2)}$  are the third rank tensors of the nonlinear quadratic susceptibility;  $\Delta k$  represents the normal component of the wave mismatch vector,

$$(I.6) \quad \Delta k = (\mathbf{k}_{\omega_{3,0}} - \mathbf{k}_{\omega_{1,0}} - \mathbf{k}_{\omega_{2,0}})_z,$$

the coordinate system  $x, y, z$  being oriented so that the  $z$ -axis is normal to the first boundary of the nonlinear medium.

The homogeneous equations (I.3) for non-perturbed amplitudes have the well-known solutions

$$(I.7) \quad A_{j,0}(t, \mathbf{r}) = A_{j,0} \left( t - \frac{\mathbf{f}_j \cdot \mathbf{r}}{u_{rgj}} \right) \quad (j = 1, 2, 3).$$

Thus, if considering the interaction of spatially unlimited plane waves, we can write the boundary conditions for sum-frequency generation in the following form

$$(I.8) \quad \begin{aligned} A_j(t, x, y, z \leq 0) &= A_{j,0} \left( t - \frac{\mathbf{f}_j \cdot \mathbf{r}}{u_{rgj}} \right) \quad \text{for } j = 1, 2, \\ A_3(t, x, y, z \leq 0) &= 0. \end{aligned}$$

Equations (I.3a–c) will be solved by means of the iterative method. For the first-step iterative solution we assume that neither of the generating radiations at  $\omega_1$  and  $\omega_2$  are perturbed by the nonlinear interaction and they obey the solution of homogeneous equation (I.7) [1, 16, 21].

The second-step iterative solution of equations (I.3a–c), together with the boundary conditions (I.8), leads to the following expression for the stochastic amplitude of generated sum-frequency wave [21]

$$(I.9) \quad A_3(t, x, y, z) = A_3^{(1)}(t, x, y, z) + A_3^{(2)}(t, x, y, z),$$

where

$$(I.9a) \quad A_3^{(1)}(t, x, y, z) =$$

$$= i\sigma_3 \int_0^z d\xi \exp(-i \Delta k \xi) A_{1,0} \left( t - \frac{\mathbf{f}_1 \cdot \mathbf{r}}{u_{rg1}} - \varepsilon_{13}z + \varepsilon_{13}\xi \right) \\ \times A_{2,0} \left( t - \frac{\mathbf{f}_2 \cdot \mathbf{r}}{u_{rg2}} - \varepsilon_{23}z + \varepsilon_{23}\xi \right)$$

and

$$(I.9b) \quad A_3^{(2)}(t, x, y, z) = \\ = -i\sigma_1\sigma_3^2 \int_0^z d\xi \int_0^\xi d\xi' \int_0^{\xi'} d\xi'' \exp[-i \Delta k(\xi - \xi' + \xi'')] \\ \times A_{2,0} \left[ t - \frac{\mathbf{f}_2 \cdot \mathbf{r}}{u_{rg2}} - \varepsilon_{23}(z - \xi) \right] A_{2,0}^* \left[ t - \frac{\mathbf{f}_2 \cdot \mathbf{r}}{u_{rg2}} - \varepsilon_{23}(z - \xi) - \varepsilon_{21}(\xi - \xi') \right] \\ \times A_{2,0} \left[ t - \frac{\mathbf{f}_2 \cdot \mathbf{r}}{u_{rg2}} - \varepsilon_{23}(z - \xi + \xi' - \xi'') - \varepsilon_{21}(\xi - \xi') \right] \\ \times A_{1,0} \left[ t - \frac{\mathbf{f}_1 \cdot \mathbf{r}}{u_{rg1}} - \varepsilon_{13}(z - \xi + \xi' - \xi'') \right] \\ - i\sigma_2\sigma_3^2 \int_0^z d\xi \int_0^\xi d\xi' \int_0^{\xi'} d\xi'' \exp[-i \Delta k(\xi - \xi' + \xi'')] \\ \times A_{1,0} \left[ t - \frac{\mathbf{f}_1 \cdot \mathbf{r}}{u_{rg1}} - \varepsilon_{13}(z - \xi) \right] A_{1,0}^* \left[ t - \frac{\mathbf{f}_1 \cdot \mathbf{r}}{u_{rg1}} - \varepsilon_{13}(z - \xi) - \varepsilon_{12}(\xi - \xi') \right] \\ \times A_{1,0} \left[ t - \frac{\mathbf{f}_1 \cdot \mathbf{r}}{u_{rg1}} - \varepsilon_{13}(z - \xi + \xi' - \xi'') - \varepsilon_{12}(\xi - \xi') \right] \\ \times A_{2,0} \left[ t - \frac{\mathbf{f}_2 \cdot \mathbf{r}}{u_{rg2}} - \varepsilon_{23}(z - \xi + \xi' - \xi'') \right],$$

$\varepsilon_{jk}$  being typical dispersion coefficients defined by

$$(I.10) \quad \varepsilon_{jk} = \frac{1}{\cos \beta_k} \left( \frac{1}{u_{rgk}} - \frac{\cos \alpha_{jk}}{u_{rgj}} \right) \quad (j, k = 1, 2, 3; \quad j \neq k),$$

$\alpha_{jk}$  are the divergence angles between two ray directions  $\mathbf{f}_j$  and  $\mathbf{f}_k$ ,  $\beta_j$  are refractive angles of the ray directions and  $\sigma_j = \gamma_j / \cos \beta_j$ .

Considering the uncorrelated sub-frequency input radiations, for which it holds that (see e.g. [41])

(I.11)

$$\langle A_{1,0}(t_1) A_{1,0}^*(t_2) A_{1,0}(t_3) A_{1,0}^*(t_4) \dots A_{2,0}(t'_1) A_{2,0}^*(t'_2) A_{2,0}(t'_3) A_{2,0}^*(t'_4) \dots \rangle = \\ = \langle A_{1,0}(t_1) A_{1,0}^*(t_2) A_{1,0}(t_3) A_{1,0}^*(t_4) \dots \rangle \langle A_{2,0}(t'_1) A_{2,0}^*(t'_2) A_{2,0}(t'_3) A_{2,0}^*(t'_4) \dots \rangle,$$

where the angle brackets denote an ensemble average, we can calculate the mean intensity of generated sum-frequency radiation in the second-step approximation of the iterative method as follows

$$(I.12) \quad \begin{aligned} \langle I_3(z) \rangle &= \left(\frac{\varepsilon_0}{\mu_0}\right)^{1/2} \frac{1}{2} n_3 \cos \delta_3 \langle A_3(z) A_3^*(z) \rangle = \\ &= \left(\frac{\varepsilon_0}{\mu_0}\right)^{1/2} \frac{1}{2} n_3 \cos \delta_3 [\langle A_3(z) A_3^*(z) \rangle^{(1)} + \langle A_3(z) A_3^*(z) \rangle^{(2)}], \end{aligned}$$

where

$$(I.12a) \quad \begin{aligned} \langle A_3(z) A_3^*(z) \rangle^{(1)} &= \sigma_3^2 \int_0^z d\xi_1 \int_0^z d\xi_2 \exp[-i \Delta k(\xi_1 - \xi_2)] \\ &\times \langle A_{1,0}(t) A_{1,0}^*[t + \varepsilon_{13}(\xi_1 - \xi_2)] \rangle \langle A_{2,0}(t) A_{2,0}^*[t + \varepsilon_{23}(\xi_1 - \xi_2)] \rangle \end{aligned}$$

and

$$(I.12b) \quad \begin{aligned} \langle A_3(z) A_3^*(z) \rangle^{(2)} &= \\ &= -\sigma_1 \sigma_3^3 \int_0^z d\xi_1 \int_0^z d\xi_2 \int_0^{\xi_2} d\xi_2' \int_0^{\xi_2'} d\xi_2'' \{ \exp[-i \Delta k(\xi_1 - \xi_2 + \xi_2' - \xi_2'')] \\ &\quad \times \langle A_{1,0}(t) A_{1,0}^*[t + \varepsilon_{13}(\xi_1 - \xi_2 + \xi_2' - \xi_2'')] \rangle \\ &\times \langle A_{2,0}(t) A_{2,0}^*[t + \varepsilon_{23}(\xi_1 - \xi_2)] A_{2,0}[t + \varepsilon_{23}(\xi_1 - \xi_2) + \varepsilon_{21}(\xi_2 - \xi_2')] \rangle \\ &\quad \times A_{2,0}^*[t + \varepsilon_{23}(\xi_1 - \xi_2 + \xi_2' - \xi_2'')] + \text{c.c.} \} \\ &- \sigma_2 \sigma_3^3 \int_0^z d\xi_1 \int_0^z d\xi_2 \int_0^{\xi_2} d\xi_2' \int_0^{\xi_2'} d\xi_2'' \{ \exp[-i \Delta k(\xi_1 - \xi_2 + \xi_2' - \xi_2'')] \\ &\quad \times \langle A_{2,0}(t) A_{3,0}^*[t + \varepsilon_{23}(\xi_1 - \xi_2 + \xi_2' - \xi_2'')] \rangle \\ &\times \langle A_{1,0}(t) A_{1,0}^*[t + \varepsilon_{13}(\xi_1 - \xi_2)] A_{1,0}[t + \varepsilon_{13}(\xi_1 - \xi_2) + \varepsilon_{12}(\xi_2 - \xi_2')] \rangle \\ &\quad \times A_{1,0}^*[t + \varepsilon_{13}(\xi_1 - \xi_2 + \xi_2' - \xi_2'')] + \text{c.c.} \}. \end{aligned}$$

The degree of coherence [41] of generated sum-frequency radiation is calculated in the first iterative approximation only,

$$(I.13) \quad \gamma_3(T, z) = \frac{\langle A_3(t, z) A_3^*(t + T, z) \rangle^{(1)}}{\langle A_3(z) A_3^*(z) \rangle^{(1)}},$$

where

$$(I.13a) \quad \begin{aligned} \langle A_3(t, z) A_3^*(t + T, z) \rangle^{(1)} &= \sigma_3^2 \int_0^z d\xi_1 \int_0^z d\xi_2 \exp[-i \Delta k(\xi_1 - \xi_2)] \\ &\times \langle A_{1,0}(t) A_{1,0}^*[t + T + \varepsilon_{13}(\xi_1 - \xi_2)] \rangle \langle A_{2,0}(t) A_{2,0}^*[t + T + \varepsilon_{23}(\xi_1 - \xi_2)] \rangle \end{aligned}$$

and  $\langle A_3(z) A_3^*(z) \rangle^{(1)}$  is given by equation (I.12a).

The spectral density of sum-frequency radiation can be obtained by means of Fourier transformation,

$$(I.14) \quad g_3(\omega, z) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \gamma_3(T, z) \exp[-i(\omega - \omega_{3,0}) T] dT.$$

From the above treatment it is clear that for explicit calculation of the mean intensity and the spectral distribution of sum-frequency radiation, which is generated in an

incoherent nonlinear optical interaction, it is necessary to know the mean values  $\langle A_{j,0}(t_1) A_{j,0}^*(t_2) \rangle$  and  $\langle A_{j,0}(t_1) A_{j,0}^*(t_2) A_{j,0}(t_3) A_{j,0}^*(t_4) \rangle$  ( $j = 1, 2$ ).

In the further treatment we shall consider the coherent input radiations, for which it holds in the classical approach that (see e.g. [41, 42])

$$(I.15a) \quad \langle A_{j,0}(t_1) A_{j,0}^*(t_2) \rangle_{\text{coh}} = \langle A_{j,0} A_{j,0}^* \rangle$$

and

$$(I.15b) \quad \langle A_{j,0}(t_1) A_{j,0}^*(t_2) A_{j,0}(t_3) A_{j,0}^*(t_4) \rangle_{\text{coh}} = \langle A_{j,0} A_{j,0}^* \rangle^2,$$

and the chaotic input radiations obeying the following factorization relations [2, 43]

$$(I.16a) \quad \langle A_{j,0}(t_1) A_{j,0}^*(t_2) \rangle_{\text{chaot}} = \langle A_{j,0} A_{j,0}^* \rangle \exp(-\Gamma_{j,0}|t_2 - t_1|)$$

and [41, 43]

$$(I.16b) \quad \begin{aligned} \langle A_{j,0}(t_1) A_{j,0}^*(t_2) A_{j,0}(t_3) A_{j,0}^*(t_4) \rangle_{\text{chaot}} &= \\ &= \langle A_{j,0}(t_1) A_{j,0}^*(t_2) \rangle \langle A_{j,0}(t_3) A_{j,0}^*(t_4) \rangle + \\ &+ \langle A_{j,0}(t_1) A_{j,0}^*(t_4) \rangle \langle A_{j,0}(t_3) A_{j,0}^*(t_2) \rangle = \\ &= \langle A_{j,0} A_{j,0}^* \rangle^2 \{ \exp[-\Gamma_{j,0}(|t_2 - t_1| + |t_4 - t_3|)] + \\ &+ \exp[-\Gamma_{j,0}(|t_2 - t_3| + |t_4 - t_1|)] \}, \end{aligned}$$

where  $\Gamma_{j,0}$  is the spectral halfwidth of the  $j$ -th chaotic mode of sub-frequency input radiation.

For lucidity, we introduce the photon fluxes, in contradistinction to the usual intensities, in our further treatment; these being defined as follows [40, 44]:

$$(I.17) \quad N_j = \frac{I_j \cos \beta_j}{\hbar \omega_j} = \left( \frac{\varepsilon_0}{\mu_0} \right)^{1/2} \frac{n_j \cos \delta_j \cos \beta_j}{2\hbar \omega_j} A_j A_j^* (j = 1, 2, 3),$$

where  $\hbar$  is Planck's constant divided by  $2\pi$ . However, it is necessary to stress that the photon fluxes represent classical quantities here.

Further, we introduce the reduced variable  $\tau$  that is defined in the following way

$$(I.18) \quad \tau = \langle N_{1,0} \rangle^{1/4} \langle N_{2,0} \rangle^{1/4} \mu z$$

and the reduced phase mismatch

$$(I.19) \quad \Delta = \frac{\Delta k}{\langle N_{1,0} \rangle^{1/4} \langle N_{2,0} \rangle^{1/4} \mu},$$

where  $\langle N_{j,0} \rangle = \langle A_{j,0} A_{j,0}^* \rangle$  ( $j = 1, 2$ ) are the mean photon fluxes of sub-frequency input radiations and  $\mu$  denotes the nonlinear coupling constant,

$$(I.20) \quad \mu = K \left[ \left( \frac{\mu_0}{\varepsilon_0} \right)^{3/2} \frac{2\hbar \omega_1 \omega_2 \omega_3}{n_1 n_2 n_3 \cos \delta_1 \cos \delta_2 \cos \delta_3 \cos \beta_1 \cos \beta_2 \cos \beta_3} \right].$$

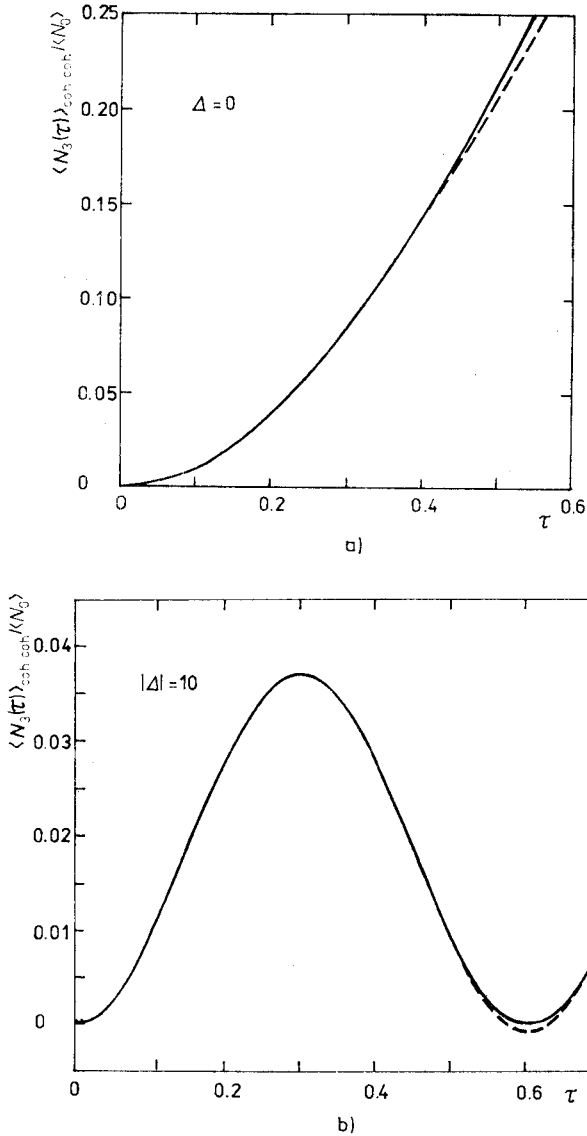


Fig. 1. Evolution of the relative mean photon flux of sum-frequency radiation  $\langle N_3(\tau) \rangle_{\text{coh coh}} / \langle N_0 \rangle$  in the sum-frequency generation with both coherent input radiations of equal mean photon fluxes ( $\langle N_{1,0} \rangle = \langle N_{2,0} \rangle = \langle N_0 \rangle$ ) for a) perfect phase matching ( $\Delta = 0$ ) and b) great phase mismatch ( $|\Delta| = 10$ ). The full lines stand for the exact solution after [40, 45] and the dashed lines correspond to the second approximation solution of the iterative method (I.23).

Besides, we define the reduced spectral half widths of input radiations

$$(I.21) \quad \eta_j = \frac{\Gamma_{j,0} |\epsilon_{j3}|}{\langle N_{1,0} \rangle^{1/4} \langle N_{2,0} \rangle^{1/4} \mu} \quad (j = 1, 2)$$

and the ratios of typical dispersion coefficients

$$(I.22) \quad \kappa_1 = \frac{\varepsilon_{12}}{\varepsilon_{13}}, \quad \kappa_2 = \frac{\varepsilon_{21}}{\varepsilon_{23}}.$$

In the case that both sub-frequency input radiations at  $\omega_1$  and  $\omega_2$  are coherent, it is evident from equations (I.12), (I.15), (I.17), (I.18), (I.19) and (I.20) that the efficiency of the process is described in the second approach of the iterative method by

$$(I.23) \quad \langle N_3(z) \rangle_{\text{coh coh}} = \langle N_{1,0} \rangle^{1/2} \langle N_{2,0} \rangle^{1/2} \frac{4}{\Delta^2} \sin^2 \left( \frac{\Delta\tau}{2} \right) - \\ - (\langle N_{1,0} \rangle + \langle N_{2,0} \rangle) \frac{4}{\Delta^2} \left[ \frac{4}{\Delta^2} \sin^2 \left( \frac{\Delta\tau}{2} \right) - \frac{\tau}{\Delta} \sin(\Delta\tau) \right],$$

which corresponds to the results of classical theory of interaction of deterministic monochromatic plane waves (cf. [40, 45]). The generated sum-frequency radiation remains coherent in the whole course of the process from the classical point of view in this case,

$$(I.24) \quad g_{3\text{coh coh}}(\omega) = \delta(\omega - \omega_{3,0}),$$

(cf. [39]).

In order to get an opinion about the accuracy of our iterative solution, we compare the second approximation solution for the mean photon flux of sum-frequency radiation (I.23) with the exact solution [40, 45] in fig. 1. It follows that the iterative solution given here represents a good approach up to the values of reduced normal distance  $\tau \approx 0.4-0.5$ . With increasing phase mismatch  $\Delta$  (decreasing efficiency of the process) (compare figs. 1a and 1b), as well as with decreasing ratio of the mean photon fluxes of input radiations  $\langle N_{2,0} \rangle / \langle N_{1,0} \rangle$  (see [21] and [39]) the extent of authentic values  $\tau$  becomes somewhat greater.

In the following treatment we shall calculate the efficiency of incoherent sum-frequency generation in terms of  $\langle N_3(\tau) \rangle$  and the spectral distribution of generated radiation  $g_3(\omega, \tau)$  for one coherent and the other chaotic input radiations, and for both chaotic input radiations, respectively.

### 3. GENERAL RESULTS

#### 3.1. Coherent and chaotic input radiations

As the first case we shall consider the incoherent sum-frequency generation with one coherent ( $\omega_1$ ) and the other chaotic ( $\omega_2$ ) sub-frequency radiations.

From equations (I.12) and (I.15-22) we can find the expression for the mean photon flux in sum-frequency radiation in the second-step approximation of the iterative method as follows:

$$(I.25) \quad \langle N_3(\tau) \rangle_{\text{coh chaot}} = \langle N_3(\tau) \rangle_{\text{coh chaot}}^{(1)} + \langle N_3(\tau) \rangle_{\text{coh chaot}}^{(2)},$$



where

$$(I.25a) \quad \langle N_3(\tau) \rangle_{\text{coh chaot}}^{(1)} = \langle N_{1,0} \rangle^{1/2} \langle N_{2,0} \rangle^{1/2} \mathcal{F}(\eta_2, \Delta, \tau)$$

and

$$(I.25b) \quad \begin{aligned} \langle N_3(\tau) \rangle_{\text{coh chaot}}^{(2)} &= -\langle N_{1,0} \rangle \mathcal{E}_1^{(c, \text{ch})}(\eta_2, \Delta, \tau) \\ &- \langle N_{2,0} \rangle [\mathcal{E}_2^{(c, \text{ch})}(\eta_2, \Delta, \tau) + \mathcal{F}_0^{(c, \text{ch})}(\eta_2, \kappa_2, \Delta, \tau)], \end{aligned}$$

with

$$(I.26) \quad \begin{aligned} \mathcal{F}(\eta, \Delta, \tau) &= \int_0^\tau dx_1 \int_0^\tau dx_2 \exp[-i\Delta(x_1 - x_2) - \eta|x_1 - x_2|] = \\ &= \frac{2\eta}{(\eta^2 + \Delta^2)} \tau + \frac{2}{(\eta^2 + \Delta^2)^2} [(\eta^2 - \Delta^2) \cos(\Delta\tau) - 2\eta\Delta \sin(\Delta\tau)] \exp(-\eta\tau) \\ &\quad - \frac{2(\eta^2 - \Delta^2)}{(\eta^2 + \Delta^2)^2}, \end{aligned}$$

$$(I.27) \quad \begin{aligned} \mathcal{E}_1^{(c, \text{ch})}(\eta, \Delta, \tau) &= \\ &= 2 \int_0^\tau dx_1 \int_0^\tau dx_2 \int_0^{x_2} dx_2' \int_0^{x_2'} dx_2'' \cos[\Delta(x_1 - x_2 + x_2' - x_2'')] \\ &\quad \times \exp(-\eta|x_1 - x_2 + x_2' - x_2''|) = \\ &= \frac{2\eta}{3(\eta^2 + \Delta^2)} \tau^3 - \frac{4}{(\eta^2 + \Delta^2)^3} \{\eta(\eta^2 - 3\Delta^2) [1 + \cos(\Delta\tau) \exp(-\eta\tau)] \\ &- \Delta(3\eta^2 - \Delta^2) \sin(\Delta\tau) \exp(-\eta\tau)\} \tau + \frac{8}{(\eta^2 + \Delta^2)^4} \{(\eta^4 - 6\eta^2\Delta^2 + \Delta^4) \\ &\times [1 - \cos(\Delta\tau) \exp(-\eta\tau)] + 4\eta\Delta(\eta^2 - \Delta^2) \sin(\Delta\tau) \exp(-\eta\tau)\}, \end{aligned}$$

$$(I.28) \quad \begin{aligned} \mathcal{E}_2^{(c, \text{ch})}(\eta, \Delta, \tau) &= \\ &= 2 \int_0^\tau dx_1 \int_0^\tau dx_2 \int_0^{x_2} dx_2' \int_0^{x_2'} dx_2'' \cos[\Delta(x_1 - x_2 + x_2' - x_2'')] \times \\ &\quad \times \exp[-\eta(|x_1 - x_2| + x_2' - x_2'')] = \\ &= \frac{2\eta^2}{(\eta^2 + \Delta^2)^2} \tau^2 - \frac{2}{(\eta^2 + \Delta^2)^3} \\ &\times \{\eta(3\eta^2 - 5\Delta^2) - 2\Delta[2\eta\Delta \cos(\Delta\tau) + (\eta^2 - \Delta^2) \sin(\Delta\tau)] \exp(-\eta\tau)\} \tau \\ &\quad + \frac{1}{(\eta^2 + \Delta^2)^4} \{7\eta^4 - 34\eta^2\Delta^2 + 7\Delta^4 + (\eta^2 + \Delta^2)^2 \exp(-2\eta\tau) \\ &- 8[(\eta^4 - 4\eta^2\Delta^2 + \Delta^4) \cos(\Delta\tau) - 3\eta\Delta(\eta^2 - \Delta^2) \sin(\Delta\tau)] \exp(-\eta\tau)\}, \end{aligned}$$

$$(I.29) \quad \begin{aligned} \mathcal{F}_0^{(c, \text{ch})}(\eta, \kappa, \Delta, \tau) &= \\ &= 2 \int_0^\tau dx_1 \int_0^\tau dx_2 \int_0^{x_2} dx_2' \int_0^{x_2'} dx_2'' \cos[\Delta(x_1 - x_2 + x_2' - x_2'')] \\ &\quad \times \exp\{-\eta[|\kappa|(x_2 - x_2') + |x_1 - (1 - \kappa)(x_2 - x_2') - x_2''|]\}; \end{aligned}$$

(i) for  $\kappa > 0$

$$\begin{aligned}
 \text{(I.29a)} \quad \mathcal{F}_0^{c,\text{ch}}(\eta, \kappa, \Delta, \tau) &= \frac{2\eta^2}{\kappa(\eta^2 + \Delta^2)^2} \tau^2 - \frac{2\eta[2(\eta^2 - \Delta^2) + \kappa^2(\eta^2 - 3\Delta^2)]}{\kappa^2(\eta^2 + \Delta^2)^3} \tau \\
 &- \frac{2}{(\eta^2 + \Delta^2)^2} \left( \eta \left\{ \frac{(\eta^2 - 3\Delta^2)}{(\eta^2 + \Delta^2)} + \frac{[\eta^2 - 3\Delta^2 - 2\kappa(\eta^2 - \Delta^2)]}{[\eta^2(1 - 2\kappa)^2 + \Delta^2]} \right\} \cos(\Delta\tau) \right. \\
 &- \Delta \left. \left\{ \frac{(3\eta^2 - \Delta^2)}{(\eta^2 + \Delta^2)} + \frac{[\eta^2(3 - 4\kappa) - \Delta^2]}{[\eta^2(1 - 2\kappa)^2 + \Delta^2]} \right\} \sin(\Delta\tau) \right) \tau \exp(-\eta\tau) \\
 &+ \frac{1}{(\eta^2 + \Delta^2)^2} \frac{([\eta^2 - 3\Delta^2 - 2\kappa(\eta^2 - \Delta^2)])}{\kappa} \\
 &+ \frac{2\{\eta^4 - 6\eta^2\Delta^2 + \Delta^4 + 4\eta^2\kappa[\kappa(\eta^2 - \Delta^2) - \eta^2 + 3\Delta^2]\}}{[\eta^2(1 - 2\kappa)^2 + \Delta^2]} \exp(-2\kappa\eta\tau) \\
 &+ \frac{1}{(\eta^2 + \Delta^2)^2} \left\{ \left( \frac{(\eta^2 - 3\Delta^2)}{\kappa(\eta^2 + \Delta^2)} - \frac{4(\eta^4 - 6\eta^2\Delta^2 + \Delta^4)}{(\eta^2 + \Delta^2)^2} + \frac{[2\kappa(\eta^2 - \Delta^2) - \eta^2 + 3\Delta^2]}{\kappa[\eta^2(1 - 2\kappa)^2 + \Delta^2]} \right. \right. \\
 &- \left. \left. \frac{2\{\eta^4 - 6\eta^2\Delta^2 + \Delta^4 + 4\eta^2\kappa[\kappa(\eta^2 - \Delta^2) - \eta^2 + 3\Delta^2]\}}{[\eta^2(1 - 2\kappa)^2 + \Delta^2]^2} \right) \cos(\Delta\tau) \right. \\
 &- \Delta \left( \frac{(3\eta^2 - \Delta^2)}{\kappa\eta(\eta^2 + \Delta^2)} - \frac{16\eta(\eta^2 - \Delta^2)}{(\eta^2 + \Delta^2)^2} - \frac{[\eta^2(3 - 4\kappa) - \Delta^2]}{\kappa\eta[\eta^2(1 - 2\kappa)^2 + \Delta^2]} \right. \\
 &- \left. \frac{8\eta(1 - \kappa)[\eta^2(1 - 2\kappa) - \Delta^2]}{[\eta^2(1 - 2\kappa)^2 + \Delta^2]^2} \right) \sin(\Delta\tau) \left. \right\} \exp(-\tau\eta) - \frac{4}{\kappa^3(\eta^2 + \Delta^2)^4} \\
 &\times [\eta^2(\eta^2 - 3\Delta^2) \cos(\kappa\Delta\tau) - \Delta\eta(3\eta^2 - \Delta^2) \sin(\kappa\Delta\tau)] \exp(-\kappa\eta\tau) \\
 &+ \frac{1}{\kappa(\eta^2 + \Delta^2)^3} \left\{ \frac{4[\eta^2(\eta^2 - 3\Delta^2) + \kappa^3(\eta^4 - 6\eta^2\Delta^2 + \Delta^4)]}{\kappa^2(\eta^2 + \Delta^2)} - \eta^2 + 3\Delta^2 \right\},
 \end{aligned}$$

(ii) for  $\kappa < 0$

$$\begin{aligned}
 \text{(I.29b)} \quad \mathcal{F}_0^{c,\text{ch}}(\eta, \kappa, \Delta, \tau) &= \frac{2\eta^2}{|\kappa|(\eta^2 + \Delta^2)^2} \tau^2 - \frac{2\eta}{\kappa^2(\eta^2 + \Delta^2)^2} \left\{ \frac{\eta^2(2 + \kappa) - \Delta^2(2 + 3\kappa^2)}{(\eta^2 + \Delta^2)} \right. \\
 &+ \left. \frac{\kappa^2[\eta^2(1 + 2|\kappa|) - \Delta^2(3 + 2|\kappa|)]}{[\eta^2(1 + 2|\kappa|)^2 + \Delta^2]} \right\} \tau - \frac{2}{(\eta^2 + \Delta^2)^3} \\
 &\times [\eta(\eta^2 - 3\Delta^2) \cos(\Delta\tau) - \Delta(3\eta^2 - \Delta^2) \sin(\Delta\tau)] \tau \exp(-\eta\tau) \\
 &+ \frac{1}{(\eta^2 + \Delta^2)^3} \left[ \frac{\eta(\eta^2 - 3\Delta^2)}{|\kappa|\eta} + \frac{4(\eta^4 - 6\eta^2\Delta^2 + \Delta^4)}{(\eta^2 + \Delta^2)} \right] \cos(\Delta\tau) \\
 &- \Delta \left[ \frac{(3\eta^2 - \Delta^2)}{|\kappa|\eta} + \frac{16\eta(\eta^2 - \Delta^2)}{(\eta^2 + \Delta^2)} \right] \sin(\Delta\tau) \left. \right\} \exp(-\eta\tau) - \frac{4}{|\kappa|^3(\eta^2 + \Delta^2)^4}
 \end{aligned}$$

$$\begin{aligned}
 & \times [\eta^2(\eta^2 - 3\Delta^2) \cos(|\kappa| \Delta\tau) - \eta\Delta(3\eta^2 - \Delta^2) \sin(|\kappa| \Delta\tau)] \exp(-|\kappa| \eta\tau) \\
 & + \frac{1}{(\eta^2 + \Delta^2)^2 [\eta^2(1 + 2|\kappa|)^2 + \Delta^2]} \left\{ \frac{\eta[\eta^2(1 + 2|\kappa|) - \Delta^2(3 + 2|\kappa|)]}{|\kappa| \eta} \right. \\
 & \quad \left. - \frac{2[\eta^4(1 + 2|\kappa|)^2 - 2\eta^2\Delta^2(3 + 6|\kappa| + 2\kappa^2) + \Delta^4]}{[\eta^2(1 + 2|\kappa|)^2 + \Delta^2]} \right\} \cos(\Delta\tau) \\
 & \quad - \Delta \left\{ \frac{[\eta^2(3 + 4|\kappa|) - \Delta^2]}{|\kappa| \eta} - \frac{8\eta[\eta^2(1 + 2|\kappa|) - \Delta^2]}{[\eta^2(1 + 2|\kappa|)^2 + \Delta^2]} \right\} \sin(\Delta\tau) \\
 & \times \exp[-(1 + 2|\kappa|) \eta\tau] + \frac{2}{(\eta^2 + \Delta^2)^2} \left\{ \frac{2[\eta^4(1 + |\kappa|^3) - 3\eta^2\Delta^2(1 + 2|\kappa|^3) + \Delta^4|\kappa|^3]}{|\kappa|^3(\eta^2 + \Delta^2)^2} \right. \\
 & \quad + \frac{[\eta^4(1 + 2|\kappa|) - 6\eta^2\Delta^2(1 + |\kappa|) + \Delta^4]}{(\eta^2 + \Delta^2)[\eta^2(1 + 2|\kappa|)^2 + \Delta^2]} \\
 & \quad \left. + \frac{[\eta^4(1 + 2|\kappa|)^2 - 2\eta^2\Delta^2(3 + 6|\kappa| + 2\kappa^2) + \Delta^4]}{[\eta^2(1 + 2|\kappa|)^2 + \Delta^2]^2} \right\}.
 \end{aligned}$$

The spectral distribution of generated sum-frequency radiation  $g_{3\text{coh chaot}}(\omega, \tau)$  can be found in the first approximation, using equations (I.9a), (I.13), (I.14), (I.15a), (I.16a), (I.18–22), (I.25a) and (I.26), in the following form:

$$\text{(I.30)} \quad g_{3\text{coh chaot}}(\Omega, \tau) = \frac{|\varepsilon_{23}|}{\langle N_{1,0} \rangle^{1/4} \langle N_{2,0} \rangle^{1/4} \mu} f_{\text{coh chaot}}(\Omega, \tau),$$

where

$$\text{(I.31)} \quad \Omega = \frac{\varepsilon_{23}}{\langle N_{1,0} \rangle^{1/4} \langle N_{2,0} \rangle^{1/4} \mu} (\omega - \omega_{3,0})$$

and

$$\text{(I.32)} \quad f_{\text{coh chaot}}(\Omega, \tau) = \frac{\eta_2 \tau^2}{\pi \mathcal{J}(\eta_2, \Delta, \tau) (\eta_2^2 + \Omega^2)} \left\{ \frac{\sin[(\Delta + \Omega) \tau/2]}{(\Delta + \Omega) \tau/2} \right\}^2,$$

$\mathcal{J}(\eta_2, \Delta, \tau)$  being given by equation (I.26).

### 3.2. Chaotic input radiations

As the second case we shall consider the incoherent sum-frequency generation with both chaotic sub-frequency input radiations at  $\omega_1$  and  $\omega_2$ .

The following expression for the mean photon flux in sum-frequency radiation was found on the basis of equations (I.12) and (I.16–22) in the second-step approximation of the iterative method:

$$\text{(I.33)} \quad \langle N_3(\tau) \rangle_{\text{chaot chaot}} = \langle N_3(\tau) \rangle_{\text{chaot chaot}}^{(1)} + \langle N_3(\tau) \rangle_{\text{chaot chaot}}^{(2)},$$

where

$$(I.33a) \quad \langle N_3(\tau) \rangle_{\text{chaot chaot}}^{(1)} = \langle N_{1,0} \rangle^{1/2} \langle N_{2,0} \rangle^{1/2} \mathcal{J}(\eta_1 + \eta_2, \Delta, \tau)$$

and

$$(I.33b) \quad \langle N_3(\tau) \rangle_{\text{chaot chaot}}^{(2)} = \\ = -\langle N_{1,0} \rangle [\mathcal{E}_0^{(\text{ch, ch})}(\eta_2, \eta_1, \Delta, \tau) + \mathcal{F}_0^{(\text{ch, ch})}(\eta_2, \eta_1, \varkappa_1, \Delta, \tau)] \\ - \langle N_{2,0} \rangle [\mathcal{E}_0^{(\text{ch, ch})}(\eta_1, \eta_2, \Delta, \tau) + \mathcal{F}_0^{(\text{ch, ch})}(\eta_1, \eta_2, \varkappa_2, \Delta, \tau)],$$

$\mathcal{J}(\eta_1 + \eta_2, \Delta, \tau)$  being given by equation (I.26) and

$$(I.34) \quad \mathcal{E}_0^{(\text{ch, ch})}(\eta_1, \eta_2, \Delta, \tau) = \\ = 2 \int_0^\tau dx_1 \int_0^\tau dx_2 \int_0^{x_2} dx_2' \int_0^{x_2'} dx_2'' \cos [\Delta(x_1 - x_2 + x_2' - x_2'')] \\ \times \exp [-\eta_1|x_1 - x_2 + x_2' - x_2''| - \eta_2(|x_1 - x_2| + x_2' - x_2'')] \\ = \frac{1}{(\eta_s^2 + \Delta^2)} \left\{ \frac{2\eta_2[\eta_s^2\eta_d + \Delta^2(\eta_2 + 3\eta_1)]}{(\eta_s^2 + \Delta^2)(\eta_d^2 + \Delta^2)} - \frac{\eta_1(\eta_s\eta_d - \Delta^2)}{\eta_2(\eta_d^2 + \Delta^2)} \right\} \tau^2 + \frac{1}{(\eta_s^2 + \Delta^2)} \\ \times \left\{ \frac{\eta_1(\eta_s\eta_d - \Delta^2)}{\eta_2^2(\eta_d^2 + \Delta^2)} - \frac{2[\eta_s^2\eta_d + \Delta^2(\eta_2 + 3\eta_1)]}{(\eta_s^2 + \Delta^2)(\eta_d^2 + \Delta^2)} - \frac{4\eta_s(\eta_s^2 - 3\Delta^2)}{(\eta_s^2 + \Delta^2)^2} \right\} \tau \\ + \frac{4}{(\eta_s^2 + \Delta^2)^3(\eta_d^2 + \Delta^2)} \{ [\eta_1\eta_s^3\eta_d + 2\eta_s\Delta^2(\eta_d^2 + \eta_1\eta_2) + \Delta^4(3\eta_1 + 2\eta_2)] \cos(\Delta\tau) \\ + \Delta[\eta_s^2(\eta_1^2 + \eta_2^2 - 2\eta_1\eta_d) + 2\eta_1\Delta^2(\eta_1 + 2\eta_2) - \Delta^4] \sin(\Delta\tau) \} \tau \exp(-\eta_s\tau) \\ + \frac{[\eta_s\eta_d^2(2\eta_2^2 + \eta_1\eta_d) + 2\Delta^2(\eta_d^3 - 2\eta_1^2\eta_2) - \eta_1\Delta^4]}{2\eta_2^3(\eta_s^2 + \Delta^2)(\eta_d^2 + \Delta^2)^2} \exp(-2\eta_2\tau) - \frac{2}{(\eta_s^2 + \Delta^2)^2} \\ \times \left( \left\{ \frac{[\eta_s^2\eta_d^2 + 2\Delta^2(\eta_2^2 - 3\eta_1^2) + \Delta^4]}{(\eta_d^2 + \Delta^2)^2} + \frac{3(\eta_s^4 - 6\eta_s^2\Delta^2 + \Delta^4)}{(\eta_s^2 + \Delta^2)^2} \right\} \cos(\Delta\tau) \right. \\ \left. + 4\Delta \left\{ \frac{\eta_1(\eta_s\eta_d + \Delta^2)}{(\eta_d^2 + \Delta^2)^2} - \frac{3\eta_s(\eta_s^2 - \Delta^2)}{(\eta_s^2 + \Delta^2)^2} \right\} \sin(\Delta\tau) \right) \exp(-\eta_s\tau) \\ - \frac{1}{(\eta_s^2 + \Delta^2)} \left\{ \frac{\eta_1(\eta_s\eta_d - \Delta^2)}{2\eta_2^3(\eta_d^2 + \Delta^2)} + \frac{[\eta_s\eta_d^2 + \Delta^2(\eta_2 - 3\eta_1)]}{\eta_2(\eta_d^2 + \Delta^2)^2} \right. \\ \left. - \frac{2[\eta_s^2\eta_d^2 + 2\Delta^2(\eta_2^2 - 3\eta_1^2) + \Delta^4]}{(\eta_s^2 + \Delta^2)(\eta_d^2 + \Delta^2)^2} - \frac{6(\eta_s^4 - 6\eta_s^2\Delta^2 + \Delta^4)}{(\eta_s^2 + \Delta^2)^3} \right\},$$

$$(I.35) \quad \mathcal{F}_0^{(\text{ch, ch})}(\eta_1, \eta_2, \varkappa, \Delta, \tau) = \\ = 2 \int_0^\tau dx_1 \int_0^\tau dx_2 \int_0^{x_2} dx_2' \int_0^{x_2'} dx_2'' \cos [\Delta(x_1 - x_2 + x_2' - x_2'')] \\ \times \exp \{ -\eta_1|x_1 - x_2 + x_2' - x_2''| - \eta_2[|\varkappa|(x_2 - x_2') \\ + |x_1 - (1 - \varkappa)(x_2 - x_2') - x_2''|] \};$$

(i) for  $\kappa > 0$

$$\begin{aligned}
 \text{(I.35a)} \quad \mathcal{F}_0^{(\text{ch, ch})}(\eta_1, \eta_2, \kappa, \Delta, \tau) = & \\
 = \frac{1}{\kappa(\eta_s^2 + \Delta^2)(\eta_d^2 + \Delta^2)} & \left\{ \frac{2\eta_2[\eta_s^2\eta_d + \Delta^2(\eta_2 + 3\eta_1)]}{(\eta_{s1}^2 + \Delta^2)} - \frac{\eta_1(\eta_s\eta_d - \Delta^2)}{\eta_2} \right\} \tau^2 \\
 + \frac{1}{\kappa^2(\eta_s^2 + \Delta^2)} & \left\{ \frac{\eta_1(\eta_s\eta_d - \Delta^2)}{\eta_2^2(\eta_d^2 + \Delta^2)} - \frac{2[\eta_s^2\eta_d + \Delta^2(\eta_2 + 3\eta_1)]}{(\eta_s^2 + \Delta^2)(\eta_d^2 + \Delta^2)} - \frac{2\eta_s(\kappa^2 + 1)(\eta_s^2 - 3\Delta^2)}{(\eta_s^2 + \Delta^2)^2} \right\} \tau \\
 - \frac{2}{(\eta_s^2 + \Delta^2)^2} & \left( \left\{ \frac{[\eta_s^2\eta_k - \Delta^2(\eta_k + 2\eta_s)]}{(\eta_k^2 + \Delta^2)} + \frac{\eta_s(\eta_s^2 - 3\Delta^2)}{(\eta_s^2 + \Delta^2)} \right\} \cos(\Delta\tau) \right. \\
 - \Delta & \left. \left\{ \frac{(3\eta_s^2 - \Delta^2)}{(\eta_s^2 + \Delta^2)} + \frac{[\eta_s(\eta_s + 2\eta_k) - \Delta^2]}{(\eta_k^2 + \Delta^2)} \right\} \sin(\Delta\tau) \right) \tau \exp(-\eta_s\tau) \\
 + \frac{1}{(\eta_s^2 + \Delta^2)} & \left\{ \frac{\eta_1(\eta_s\eta_d - \Delta^2)}{2\kappa^3\eta_2^3(\eta_d^2 + \Delta^2)} + \frac{[\eta_s^2\eta_k - \Delta^2(3\eta_s - 2\kappa\eta_2)]}{\kappa\eta_2(\eta_s^2 + \Delta^2)(\eta_k^2 + \Delta^2)} \right. \\
 + \frac{2[\eta_s^2\eta_k^2 - \Delta^2(\eta_s^2 + \eta_k^2 + 4\eta_s\eta_k) + \Delta^4]}{(\eta_s^2 + \Delta^2)(\eta_k^2 + \Delta^2)^2} & \left. \right\} \exp(-2\kappa\eta_2\tau) \\
 + \frac{1}{(\eta_s^2 + \Delta^2)^2} & \left\{ \frac{\eta_s(\eta_s^2 - 3\Delta^2)}{\kappa\eta_2(\eta_s^2 + \Delta^2)} - \frac{4(\eta_s^4 - 6\eta_s^2\Delta^2 + \Delta^4)}{(\eta_s^2 + \Delta^2)^2} \right. \\
 - \frac{[\eta_s^2\eta_k - \Delta^2(2\eta_s + \eta_k)]}{\kappa\eta_2(\eta_k^2 + \Delta^2)} & - \frac{2[\eta_s^2\eta_k^2 - \Delta^2(\eta_s^2 + \eta_k^2 + 4\eta_s\eta_k) + \Delta^4]}{(\eta_k^2 + \Delta^2)^2} \left. \right\} \cos(\Delta\tau) \\
 + \Delta & \left\{ \frac{16\eta_s(\eta_s^2 - \Delta^2)}{(\eta_s^2 + \Delta^2)^2} - \frac{(3\eta_s^2 - \Delta^2)}{\kappa\eta_2(\eta_s^2 + \Delta^2)} + \frac{[\eta_s(\eta_s + 2\eta_k) - \Delta^2]}{\kappa\eta_2(\eta_k^2 + \Delta^2)} \right. \\
 + \frac{4(\eta_s + \eta_k)(\eta_s\eta_k - \Delta^2)}{(\eta_k^2 + \Delta^2)^2} & \left. \right\} \sin(\Delta\tau) \exp(-\eta_s\tau) - \frac{2}{\kappa^3(\eta_s^2 + \Delta^2)^3} \\
 \times \left( \left[ \frac{(\eta_s^4 - 6\eta_s^2\Delta^2 + \Delta^4)}{(\eta_s^2 + \Delta^2)} + \frac{(\eta_s^3\eta_d + 6\eta_s\eta_1\Delta^2 - \Delta^4)}{(\eta_d^2 + \Delta^2)} \right] \right. & \left. \cos(\kappa\Delta\tau) \right) \\
 - 2\Delta & \left\{ \frac{2\eta_s(\eta_s^2 - \Delta^2)}{(\eta_s^2 + \Delta^2)} - \frac{[\eta_s^2(2\eta_1 - \eta_2) - \Delta^2(2\eta_1 + \eta_2)]}{(\eta_d^2 + \Delta^2)} \right\} \sin(\kappa\Delta\tau) \exp(-\kappa\eta_s\tau) \\
 + \frac{1}{\kappa(\eta_s^2 + \Delta^2)} & \left[ \frac{2(2\kappa^3 + 1)(\eta_s^4 - 6\eta_s^2\Delta^2 + \Delta^4)}{\kappa^2(\eta_s^2 + \Delta^2)^3} - \frac{\eta_1(\eta_s\eta_d - \Delta^2)}{2\kappa^2\eta_2^3(\eta_d^2 + \Delta^2)} \right. \\
 + \frac{2(\eta_s^3\eta_d + 6\eta_s\eta_1\Delta^2 - \Delta^4)}{\kappa^2(\eta_s^2 + \Delta^2)^2(\eta_d^2 + \Delta^2)} & - \left. \frac{\eta_s(\eta_s^2 - 3\Delta^2)}{\eta_2(\eta_s^2 + \Delta^2)^2} \right],
 \end{aligned}$$

(ii) for  $\kappa < 0$

$$\begin{aligned}
 \text{(I.35b)} \quad \mathcal{F}_0^{\text{(ch, ch)}}(\eta_1, \eta_2, \kappa, \Delta, \tau) = & \\
 = \frac{1}{|\kappa|(\eta_s^2 + \Delta^2)(\eta_d^2 + \Delta^2)} & \left\{ \frac{2\eta_2[\eta_s^2\eta_d + \Delta^2(\eta_2 + 3\eta_1)]}{(\eta_s^2 + \Delta^2)} - \frac{\eta_1(\eta_s\eta_d - \Delta^2)}{\eta_2} \right\} \tau^2 \\
 + \frac{1}{\kappa^2(\eta_s^2 + \Delta^2)} & \left\{ \frac{\eta_1(\eta_s\eta_d - \Delta^2)}{\eta_s^2(\eta_d^2 + \Delta^2)} - \frac{2[\eta_s^2\eta_d + \Delta^2(\eta_2 + 3\eta_1)]}{(\eta_s^2 + \Delta^2)(\eta_d^2 + \Delta^2)} - \frac{2\eta_s(\kappa^2 + 1)(\eta_s^2 - 3\Delta^2)}{(\eta_s^2 + \Delta^2)^2} \right. \\
 - \frac{2\kappa^2[\eta_s^2\eta_k - \Delta^2(2\eta_s + \eta_k)]}{(\eta_s^2 + \Delta^2)(\eta_k^2 + \Delta^2)} & \left. \right\} \tau - \frac{2}{(\eta_s^2 + \Delta^2)^3} [\eta_s(\eta_s^2 - 3\Delta^2) \cos(\Delta\tau) \\
 - \Delta(3\eta_s^2 - \Delta^2) \sin(\Delta\tau)] \tau \exp(-\eta_s\tau) & + \frac{\eta_1(\eta_s\eta_d - \Delta^2)}{2|\kappa|^3 \eta_2^3(\eta_s^2 + \Delta^2)(\eta_d^2 + \Delta^2)} \exp(-2|\kappa| \eta_2\tau) \\
 - \frac{1}{(\eta_s^2 + \Delta^2)^3} & \left\{ \left[ \frac{\eta_s(\eta_s^2 - 3\Delta^2)}{|\kappa| \eta_2} + \frac{4(\eta_s^4 - 6\eta_s^2\Delta^2 + \Delta^4)}{(\eta_s^2 + \Delta^2)} \right] \cos(\Delta\tau) \right. \\
 - \Delta \left[ \frac{16\eta_s(\eta_s^2 - \Delta^2)}{(\eta_s^2 + \Delta^2)} + \frac{(3\eta_s^2 - \Delta^2)}{|\kappa| \eta_2} \right] & \left. \sin(\Delta\tau) \right\} \exp(-\eta_s\tau) \\
 - \frac{2}{|\kappa|^3(\eta_s^2 + \Delta^2)^3} & \left\{ \left[ \frac{(\eta_s^4 - 6\eta_s^2\Delta^2 + \Delta^4)}{(\eta_s^2 + \Delta^2)} + \frac{(\eta_s^3\eta_d + 6\eta_1\eta_s\Delta^2 - \Delta^4)}{(\eta_d^2 + \Delta^2)} \right] \cos(|\kappa| \Delta\tau) \right. \\
 - 2\Delta \left[ \frac{\eta_s^2(\eta_2 - 2\eta_1) + \Delta^2(\eta_2 + 2\eta_1)}{(\eta_d^2 + \Delta^2)} + \frac{2\eta_s(\eta_s^2 - \Delta^2)}{(\eta_s^2 + \Delta^2)} \right] & \left. \sin(|\kappa| \Delta\tau) \right\} \exp(-|\kappa| \eta_s\tau) \\
 + \frac{1}{(\eta_s^2 + \Delta^2)^2(\eta_k^2 + \Delta^2)} & \left\{ \left[ \frac{\eta_s^2\eta_k - \Delta^2(2\eta_s + \eta_k)}{|\kappa| \eta_2} \right. \right. \\
 - \frac{2[\eta_s^2\eta_k^2 - \Delta^2(\eta_s^2 + \eta_k^2 + 4\eta_s\eta_k) + \Delta^4]}{(\eta_k^2 + \Delta^2)} & \left. \left. \right\} \cos(\Delta\tau) \\
 + \Delta \left\{ \frac{8(\eta_s + |\kappa| \eta_1)(\eta_s\eta_k - \Delta^2)}{(\eta_k^2 + \Delta^2)} - \frac{[\eta_s(\eta_s + 2\eta_k) - \Delta^2]}{|\kappa| \eta_2} \right\} & \sin(\Delta\tau) \right\} \exp(-\eta_k\tau) \\
 + \frac{1}{(\eta_s^2 + \Delta^2)} & \left\{ \frac{2(2|\kappa|^3 + 1)(\eta_s^4 - 6\eta_s^2\Delta^2 + \Delta^4)}{|\kappa|^3(\eta_s^2 + \Delta^2)^3} + \frac{2(\eta_s^3\eta_d + 6\eta_1\eta_s\Delta^2 - \Delta^4)}{|\kappa|^3(\eta_s^2 + \Delta^2)^2(\eta_d^2 + \Delta^2)} \right. \\
 - \frac{\eta_1(\eta_s\eta_d - \Delta^2)}{2|\kappa|^3 \eta_2^3(\eta_d^2 + \Delta^2)} + \frac{2[\eta_s^3\eta_k - 3\eta_s(\eta_s + \eta_k)\Delta^2 + \Delta^4]}{(\eta_s^2 + \Delta^2)^2(\eta_k^2 + \Delta^2)} & \\
 \left. + \frac{2[\eta_s^2\eta_k^2 - \Delta^2(\eta_s^2 + \eta_k^2 + 4\eta_s\eta_k) + \Delta^4]}{(\eta_s^2 + \Delta^2)(\eta_k^2 + \Delta^2)^2} \right\}, &
 \end{aligned}$$

where  $\eta_s = \eta_1 + \eta_2$ ,  $\eta_d = \eta_2 - \eta_1$ ,  $\eta_k = (1 - 2\kappa)\eta_2 + \eta_1$ .

The spectral distribution of generated sum-frequency radiation  $g_{3\text{chaotchaot}}(\omega, \tau)$  was found in the first approximation of the iterative solution, using equations (I.9a), (I.13), (I.14), (I.16a), (I.18–22), (I.26) and (I.33a), to be:

(i) for  $|\varepsilon_{13}| \geq |\varepsilon_{23}|$

$$(I.36a) \quad g_{3\text{chaotchaot}}(\Omega, \tau) = \frac{|\varepsilon_{13}|}{\langle N_{1,0} \rangle^{1/4} \langle N_{2,0} \rangle^{1/4} \mu} f_{\text{chaotchaot}}(\Omega, \tau)$$

and

(ii) for  $|\varepsilon_{13}| \leq |\varepsilon_{23}|$

$$(I.36b) \quad g_{3\text{chaotchaot}}(\Omega, \tau) = \frac{|\varepsilon_{23}|}{\langle N_{1,0} \rangle^{1/4} \langle N_{2,0} \rangle^{1/4} \mu} f_{\text{chaotchaot}}(\Omega, \tau),$$

where

(i) for  $|\varepsilon_{13}| \geq |\varepsilon_{23}|$

$$(I.37a) \quad \Omega = \frac{|\varepsilon_{13}|}{\langle N_{1,0} \rangle^{1/4} \langle N_{2,0} \rangle^{1/4} \mu} (\omega - \omega_{3,0})$$

and

(ii) for  $|\varepsilon_{13}| \leq |\varepsilon_{23}|$

$$(I.37b) \quad \Omega = \frac{|\varepsilon_{23}|}{\langle N_{1,0} \rangle^{1/4} \langle N_{2,0} \rangle^{1/4} \mu} (\omega - \omega_{3,0});$$

$$(I.38) \quad f_{\text{chaotchaot}}(\Omega, \tau) = \frac{\Phi(\eta_1, \eta_2, m, \Delta, \Omega, \tau)}{2\pi \mathcal{J}(\eta_s, \Delta, \tau)},$$

$\mathcal{J}(\eta_s, \Delta, \tau)$  being given by equation (I.26) and  $\Phi(\eta_1, \eta_2, m, \Delta, \Omega, \tau)$  is defined as follows:

(i) for  $|\varepsilon_{13}| \geq |\varepsilon_{23}|$

$$(I.39a) \quad \begin{aligned} \Phi(\eta_1, \eta_2, m, \Delta, \Omega, \tau) = \\ = 2 \operatorname{Re} \int_0^\infty d\xi \exp(-i\Omega\xi) \int_0^\tau dx_1 \int_0^\tau dx_2 \exp[-i\Delta(x_1 - x_2) \\ - \eta_1 |\operatorname{sgn}(\varepsilon_{13}) \xi + x_2 - x_1| - \eta_2 |\operatorname{sgn}(\varepsilon_{23}) (1+m)\xi + x_2 - x_1|] \end{aligned}$$

and

(ii) for  $|\varepsilon_{13}| \leq |\varepsilon_{23}|$

$$(I.39b) \quad \begin{aligned} \Phi(\eta_1, \eta_2, m, \Delta, \Omega, \tau) = \\ = 2 \operatorname{Re} \int_0^\infty d\xi \exp(-i\Omega\xi) \int_0^\tau dx_1 \int_0^\tau dx_2 \exp[-i\Delta(x_1 - x_2) \\ - \eta_1 |\operatorname{sgn}(\varepsilon_{13}) (1+m)\xi + x_2 - x_1| - \eta_2 |\operatorname{sgn}(\varepsilon_{23}) \xi + x_2 - x_1|], \end{aligned}$$

where  $\eta_s = \eta_1 + \eta_2$ ,  $\eta_d = \eta_2 - \eta_1$  and

(i) for  $|\varepsilon_{13}| \geq |\varepsilon_{23}|$

$$(I.40a) \quad m = \frac{|\varepsilon_{13}| - |\varepsilon_{23}|}{|\varepsilon_{23}|},$$

(ii) for  $|\varepsilon_{13}| \leq |\varepsilon_{23}|$

$$(I.40b) \quad m = \frac{|\varepsilon_{23}| - |\varepsilon_{13}|}{|\varepsilon_{13}|}.$$

The calculation of integrals on the right-hand sides of equations (I.39) leads to the following expressions for  $\Phi(\eta_1, \eta_2, m, \Delta, \Omega, \tau)$  in the individual cases:

a) for  $\varepsilon_{13} > 0, \varepsilon_{23} > 0$  and  $\varepsilon_{13} \geq \varepsilon_{23}$

$$(I.41a) \quad \Phi(\eta_1, \eta_2, m, \Delta, \Omega, \tau) = \frac{2[(\eta_s^2 - \Delta^2)(\eta_s + m\eta_2) + 2\Delta\Omega\eta_s]}{(\eta_s^2 + \Delta^2)^2 [(\eta_s + m\eta_2)^2 + \Omega^2]} \cos \alpha$$

$$\times \left\{ \exp(-m\eta_2\tau) \cos [(\Omega + \Delta)\tau - \alpha] - 2 \cos \alpha + \exp\left[-\frac{m\eta_1}{(1+m)}\tau\right] \right.$$

$$\times \cos \left[ \left( \frac{\Omega}{1+m} + \Delta \right) \tau + \alpha \right] \left. \right\} + \frac{4\eta_1\{\eta_s\eta_d - \Delta^2\} [m^2\eta_2^2 - (\Omega + \Delta)^2] + 4m\Delta\eta_2^2(\Omega + \Delta)}{(\eta_s^2 + \Delta^2)(\eta_d^2 + \Delta^2) [m^2\eta_2^2 + (\Omega + \Delta)^2]^2 \cos \beta}$$

$$\times \{ \cos \beta - \exp(-m\eta_2\tau) \cos [(\Omega + \Delta)\tau + \beta] \}$$

$$- \frac{2[(\eta_d^2 - \Delta^2)(\eta_d + m\eta_2) + 2\Delta\eta_d\Omega]}{(\eta_d^2 + \Delta^2)^2 [(\eta_d + m\eta_2)^2 + \Omega^2]} \cos \zeta \left\{ \exp(-m\eta_2\tau) \cos [(\Omega + \Delta)\tau - \zeta] \right.$$

$$\left. - \exp\left[-\frac{m\eta_1}{(1+m)}\tau\right] \cos \left[ \left( \frac{\Omega}{1+m} + \Delta \right) \tau - \zeta \right] \right\}$$

$$+ \frac{8\eta_2 A \{ \cos v - \exp[-m\eta_1\tau/(1+m)] \cos [(\Omega/(1+m) + \Delta)\tau + v] \}}{(\eta_s^2 + \Delta^2)^2 (\eta_d^2 + \Delta^2)^2 \{ m^2\eta_1^2 + [\Omega + (1+m)\Delta]^2 \} \cos v}$$

$$+ \frac{4(1+m)\eta_2 B \{ \cos \vartheta - \exp[-m\eta_1\tau/(1+m)] \cos [(\Omega/(1+m) + \Delta)\tau + \vartheta] \}}{(\eta_s^2 + \Delta^2)(\eta_d^2 + \Delta^2) \{ m^2\eta_1^2 + [\Omega + (1+m)\Delta]^2 \}^2 \cos \vartheta}$$

$$- \frac{8\eta_1 D \{ \cos \lambda - \exp(-m\eta_2\tau) \cos [(\Omega + \Delta)\tau + \lambda] \}}{(\eta_s^2 + \Delta^2)^2 (\eta_d^2 + \Delta^2)^2 [m^2\eta_2^2 + (\Omega + \Delta)^2] \cos \lambda} - \frac{4\eta_1\eta_2\tau}{(\eta_s^2 + \Delta^2)(\eta_d^2 + \Delta^2)}$$

$$\times \left( \frac{[m(\eta_s\eta_d - \Delta^2) + 2\Delta(\Omega + \Delta)]}{[m^2\eta_2^2 + (\Omega + \Delta)^2]} - \frac{\{m(\eta_s\eta_d + \Delta^2) + 2\Delta[\Omega + (1+m)\Delta]\}}{\{m^2\eta_1^2 + [\Omega + (1+m)\Delta]^2\}} \right),$$

b) for  $\varepsilon_{13} > 0, \varepsilon_{23} > 0$  and  $\varepsilon_{13} \leq \varepsilon_{23}$  the expression for  $\Phi(\eta_1, \eta_2, m, \Delta, \Omega, \tau)$  is given by equation (I.41a) where we interchange  $\eta_1$  and  $\eta_2$ :  $\eta_1 \rightleftharpoons \eta_2$ ,

c) for  $\varepsilon_{13} < 0, \varepsilon_{23} < 0$  and  $|\varepsilon_{13}| \geq |\varepsilon_{23}|$  the expression for  $\Phi(\eta_1, \eta_2, m, \Delta, \Omega, \tau)$  is given by equation (I.41a) where we replace  $\Delta$  by  $-\Delta$ :  $\Delta \rightarrow -\Delta$ ,



d) for  $\varepsilon_{13} < 0$ ,  $\varepsilon_{23} < 0$  and  $|\varepsilon_{13}| \leq |\varepsilon_{23}|$  the expression for  $\Phi(\eta_1, \eta_2, m, \Delta, \Omega, \tau)$  is given by equation (I.41a) where we interchange  $\eta_1$  and  $\eta_2$ :  $\eta_1 \rightleftharpoons \eta_2$  and replace  $\Delta$  by  $-\Delta$ :  $\Delta \rightarrow -\Delta$ ,

e) for  $\varepsilon_{13} > 0$ ,  $\varepsilon_{23} < 0$  and  $|\varepsilon_{13}| \geq |\varepsilon_{23}|$

$$\begin{aligned}
 \text{(I.41b)} \quad \Phi(\eta_1, \eta_2, m, \Delta, \Omega, \tau) &= \frac{2[(\eta_s^2 - \Delta^2)(\eta_d + m\eta_2) - 2\Delta\eta_s\Omega]}{(\eta_s^2 + \Delta^2)^2 [(\eta_d + m\eta_2)^2 + \Omega^2]} \cos \varphi \\
 &\times \left\{ \cos \varphi - \exp \left[ \left( \frac{\eta_d + m\eta_2}{1 + m} \right) \tau \right] \cos \left[ \frac{\Omega}{(1 + m)} \tau - \varphi \right] + \exp \left[ - \left( \frac{2 + m}{1 + m} \right) \eta_1 \tau \right] \right. \\
 &\times \cos \left[ \left( \frac{\Omega}{1 + m} - \Delta \right) \tau - \varphi \right] - \exp [-(2 + m) \eta_2 \tau] \cos [(\Omega + \Delta) \tau + \varphi] \left. \right\} \\
 &- \frac{2[(\eta_s + m\eta_2)(\eta_d^2 - \Delta^2) - 2\Delta\eta_d\Omega]}{(\eta_d^2 + \Delta^2)^2 [(\eta_s + m\eta_2)^2 + \Omega^2]} \cos \varrho \left\{ \cos \varrho + \exp \left[ - \left( \frac{\eta_s + m\eta_2}{1 + m} \right) \tau \right] \right. \\
 &\times \cos \left[ \frac{\Omega}{(1 + m)} \tau - \varrho \right] - \exp \left[ - \left( \frac{2 + m}{1 + m} \right) \eta_1 \tau \right] \cos \left[ \left( \frac{\Omega}{1 + m} - \Delta \right) \tau - \varrho \right] \\
 &\quad \left. - \exp [-(2 + m) \eta_2 \tau] \cos [(\Omega + \Delta) \tau - \varrho] \right\} \\
 &- \frac{4\eta_1 G \{ \cos \chi - \exp [-(2 + m) \eta_2 \tau] \cos [(\Omega + \Delta) \tau - \chi] \}}{(\eta_s^2 + \Delta^2)(\eta_d^2 + \Delta^2) [(2 + m)^2 \eta_2^2 + (\Omega + \Delta)^2] \cos \chi} \\
 &+ \frac{8\eta_1 P \{ \cos \xi - \exp [-(2 + m) \eta_2 \tau] \cos [(\Omega + \Delta) \tau - \xi] \}}{(\eta_s^2 + \Delta^2)^2 (\eta_d^2 + \Delta^2)^2 [(2 + m)^2 \eta_2^2 + (\Omega + \Delta)^2] \cos \xi} \\
 &+ \frac{4\eta_1 \eta_2 \tau \{ (2 + m)(\eta_s \eta_d + \Delta^2) - 2\Delta[\Omega - (1 + m)\Delta] \}}{(\eta_s^2 + \Delta^2)(\eta_d^2 + \Delta^2) \{ (2 + m)^2 \eta_1^2 + [\Omega - (1 + m)\Delta]^2 \} \cos \psi} \\
 &\times \left\{ \cos \psi - \exp \left[ - \left( \frac{2 + m}{1 + m} \right) \eta_1 \tau \right] \cos \left[ \left( \frac{\Omega}{1 + m} - \Delta \right) \tau - \psi \right] \right\} \\
 &- \frac{4\eta_1 \eta_2 \tau [(2 + m)(\eta_s \eta_d - \Delta^2) - 2\Delta(\Omega + \Delta)]}{(\eta_s^2 + \Delta^2)(\eta_d^2 + \Delta^2) [(2 + m)^2 \eta_2^2 + (\Omega + \Delta)^2]},
 \end{aligned}$$

f) for  $\varepsilon_{13} > 0$ ,  $\varepsilon_{23} < 0$  and  $|\varepsilon_{13}| \leq |\varepsilon_{23}|$  the expression for  $\Phi(\eta_1, \eta_2, m, \Delta, \Omega, \tau)$  is given by equation (I.41b) if we interchange  $\eta_1$  and  $\eta_2$ :  $\eta_1 \rightleftharpoons \eta_2$ ,

g) for  $\varepsilon_{13} < 0$ ,  $\varepsilon_{23} > 0$  and  $|\varepsilon_{13}| \geq |\varepsilon_{23}|$  the expression for  $\Phi(\eta_1, \eta_2, m, \Delta, \Omega, \tau)$  is given by equation (I.41b) if we replace  $\Delta$  by  $-\Delta$ :  $\Delta \rightarrow -\Delta$ ,

h) for  $\varepsilon_{13} < 0$ ,  $\varepsilon_{23} > 0$  and  $|\varepsilon_{13}| \leq |\varepsilon_{23}|$  the expression for  $\Phi(\eta_1, \eta_2, m, \Delta, \Omega, \tau)$  is given by equation (I.41b) if we exchange  $\eta_1$  and  $\eta_2$ :  $\eta_1 \rightleftharpoons \eta_2$  and replace  $\Delta$  by  $-\Delta$ :  $\Delta \rightarrow -\Delta$ .

The following notation has been used here:

$$\alpha = \operatorname{arctg} \left[ \frac{2\Delta\eta_s(\eta_s + m\eta_2) - \Omega(\eta_s^2 - \Delta^2)}{(\eta_s^2 - \Delta^2)(\eta_s + m\eta_2) + 2\Delta\Omega\eta_s} \right],$$

$$\beta = \operatorname{arctg} \left( \frac{2\eta_2 \{ \Delta [ m^2\eta_2^2 - (\Omega + \Delta)^2 ] - m(\Omega + \Delta)(\eta_s\eta_d - \Delta^2) \}}{(\eta_s\eta_d - \Delta^2) [ (\Omega + \Delta)^2 - m^2\eta_2^2 ] - 4m\Delta\eta_2^2(\Omega + \Delta)} \right),$$

$$\zeta = \operatorname{arctg} \left[ \frac{\Omega(\Delta^2 - \eta_d^2) + 2\Delta\eta_d(\eta_d + m\eta_2)}{(\eta_d^2 - \Delta^2)(\eta_d + m\eta_2) + 2\Delta\eta_d\Omega} \right],$$

$$\vartheta = \operatorname{arctg} (2\eta_1 \{ \Delta [ m^2\eta_1^2 - (\Omega + \Delta + m\Delta)^2 ] - m(\Omega + \Delta + m\Delta)(\eta_s\eta_d + \Delta^2) \} B^{-1}),$$

$$\lambda = \operatorname{arctg} (\eta_2 \{ 4\Delta(\Delta^2 - \eta_s\eta_d)(m\eta_2^2 - \Delta(\Omega + \Delta)) + (\Omega + \Delta + m\Delta) \\ \times [ (\Delta^2 - \eta_s\eta_d)^2 - 4\eta_2\Delta^2 ] \} D^{-1}),$$

$$\nu = \operatorname{arctg} (\eta_1 \{ (\Omega + \Delta) [ (\eta_s\eta_d + \Delta^2)^2 - 4\eta_1^2\Delta^2 ] \\ - 4\Delta(\eta_s\eta_d + \Delta^2) [ m\eta_1^2 + \Delta(\Omega + \Delta + m\Delta) ] \} A^{-1}),$$

$$\xi = \operatorname{arctg} (\eta_2 \{ 4\Delta(\Delta^2 - \eta_s\eta_d) [ (2 + m)\eta_2^2 + \Delta(\Omega + \Delta) ] \\ - [ \Omega - (1 + m)\Delta ] [ (\eta_s\eta_d - \Delta^2)^2 - 4\eta_2^2\Delta^2 ] \} P^{-1}),$$

$$\varrho = \operatorname{arctg} \left[ \frac{\Omega(\Delta^2 - \eta_d^2) - 2\Delta\eta_d(\eta_s + m\eta_2)}{(\eta_s + m\eta_2)(\eta_d^2 - \Delta^2) - 2\Delta\eta_d\Omega} \right],$$

$$\varphi = \operatorname{arctg} \left[ \frac{2\Delta\eta_s(\eta_d + m\eta_2) + \Omega(\eta_s^2 - \Delta^2)}{(\eta_d + m\eta_2)(\eta_s^2 - \Delta^2) - 2\Delta\eta_s\Omega} \right],$$

$$\chi = \operatorname{arctg} (2\eta_2 \{ \Delta [ (2 + m)^2\eta_2^2 - (\Omega + \Delta)^2 ] + (2 + m)(\eta_s\eta_d - \Delta^2)(\Omega + \Delta) \} G^{-1}),$$

$$\psi = \operatorname{arctg} \left( \frac{2\Delta(2 + m)\eta_1^2 + (\eta_s\eta_d + \Delta^2) [ \Omega - (1 + m)\Delta ]}{\eta_1 \{ 2\Delta [ \Omega - (1 + m)\Delta ] - (2 + m)(\eta_s\eta_d + \Delta^2) \}} \right),$$

$$A = [ (\eta_s\eta_d + \Delta^2)^2 - 4\eta_1^2\Delta^2 ] \{ m\eta_1^2 + \Delta [ \Omega + (1 + m)\Delta ] \} \\ + 4\Delta\eta_1^2(\eta_s\eta_d + \Delta^2)(\Omega + \Delta),$$

$$B = (\eta_s\eta_d + \Delta^2) \{ [ \Omega + (1 + m)\Delta ]^2 - m^2\eta_1^2 \} - 4\eta_1^2m\Delta [ \Omega + (1 + m)\Delta ],$$

$$D = [ (\eta_s\eta_d - \Delta^2)^2 - 4\Delta^2\eta_2^2 ] [ m\eta_2^2 - \Delta(\Omega + \Delta) ] \\ + 4\Delta\eta_2^2(\eta_s\eta_d - \Delta^2) [ \Omega + (1 + m)\Delta ],$$

$$G = (\eta_s\eta_d - \Delta^2) [ (\Omega + \Delta)^2 - (2 + m)^2\eta_2^2 ] + 4\Delta\eta_2^2(2 + m)(\Omega + \Delta),$$

$$P = [ (\eta_s\eta_d - \Delta^2)^2 - 4\Delta^2\eta_2^2 ] [ (2 + m)\eta_2^2 + \Delta(\Omega + \Delta) ] \\ - 4\Delta\eta_2^2(\eta_s\eta_d - \Delta^2) [ \Omega - (1 + m)\Delta ].$$

From the above treatment it is clear that both the efficiency of incoherent sum-frequency generation and the spectral distribution of generated radiation depend upon many parameters. In order to get an opinion about the evolution of the field

in an incoherent nonlinear optical process it is necessary to make certain simplifications and to discuss particularly special cases that roughly approximate the situation in real experiments. This will be performed in the following three papers.

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