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QUANTUM STATISTICAL PROPERTIES OF HIGHER HARMONICS AND SUBHARMONICS

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Using the power series solution of the Heisenberg–Langevin equations as well as the iterative solution of the generalized Fokker–Planck equation we study the statistical properties of higher harmonics and subharmonics generated in an optical non-linear process. A combined method using the Laplace transformation and integration by parts is proposed to solve the Heisenberg–Langevin equations, which provides a fuller description of the lossy mechanism in single iterations and the power series solution follows as a special case. In the approximations used the quantum statistics is described by the superposition of coherent and chaotic fields. While in the higher-harmonics generation the pumping radiation is getting noise and higher harmonics have a tendency to the coherent, in the subharmonics generation the pumping radiation has tendency to be coherent and the subharmonics are getting the noise. Some qualitative differences between the second and higher subharmonics are found. Both the above approaches are found to be equivalent up to the second iteration although the generalized Fokker–Planck equation approach provides a more compact solution. Making use of the phase diffusion model of laser light, it is shown that the spectrum of the k-th harmonic is k^2 broader than that of the pumping radiation while the spectrum of the k-th subharmonic is k^2 narrower.

1. Introduction

In the present paper we deal with the quantum statistical properties of higher harmonically and subharmonically generated radiation with special attention to the generation of the second harmonic and subharmonic. This paper can be considered as a continuation of papers [1, 2]. We adopt both the Heisenberg (the Heisenberg–Langevin equations,

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quantum characteristic functions and quasi-distributions) and Schrödinger (generalized Fokker–Planck equation) approaches. We show that the solution of the Heisenberg–Langevin equations (the lossy mechanism is taken into account in the usual quantum mechanical way) in powers of the interaction time $t = z/c$ (z being the distance travelled in the active medium and c light velocity in the medium) corresponds to the iterative solution of the generalized Fokker-Planck equation. Further we propose a new method for solving the Heisenberg–Langevin equations based on combining the Laplace transform method and integration by parts. It provides full reservoir contribution in an approximate solution and the power series solution follows as a special case. The method is completely described in [3]. As has been shown in [1, 2], in the higher-harmonics generation process the pumping radiation is getting quantum noise while generated higher harmonics have a tendency to be coherent. We show here on the contrary that for the generation of subharmonics the pumping radiation has a tendency to be coherent while subharmonics are getting the quantum noise similarly as in the case of a general scattering process (parametric amplification process) discussed in [1, 2]. In this case there is some qualitative difference between the second and higher order phenomena as has been also found in [4]. Finally, making use of the realistic diffusion phase model of laser light we show that if the pumping radiation has a Lorentzian spectrum of halfwidth Γ_1, then the halfwidth of the spectrum of the k-th harmonic is $k^2\Gamma_1$ (this has been obtained for $k = 2$ in [5] through a calculation of the Glauber–Sudarshan quasi-distribution) while the halfwidth of the spectrum of the k-th subharmonic is Γ_1/k^2. In the first case we have an example of a fully coherent field with an arbitrary spectral composition which is non-stationary, in the second case we have an example of noise field with improving second-order coherence.

2. *Heisenberg–Langevin approach*

The above non-linear optical processes are described by the Hamiltonian

$$H = \hbar\omega_1 a_1^+ a_1 + \hbar\omega_2 a_2^+ a_2 + \hbar g a_1^k a_2^+ + \hbar g^* a_1^{+k} a_2, \qquad k \geqslant 2, \tag{2.1}$$

where $\hbar$ is the Planck constant divided by 2π, ω_1 and ω_2 are frequencies of the pumping radiation (k-th subharmonic) mode 1 and k-th harmonic (pumping radiation) mode 2, respectively, g is a coupling constant and a_j and a_j^+ are the annihilation and creation operators of a photon in the j-th mode ($j = 1, 2$) respectively. The resonance frequency condition

$$k\omega_1 = \omega_2 \tag{2.2}$$

is assumed.

The lossy mechanism is described by the Hamiltonian

$$H_{\text{ad}} = \sum_{j=1}^{2} \sum_{l} (\hbar\psi_l^{(j)} b_l^{(j)+} b_l^{(j)} + \hbar\kappa_{jl} a_j^+ b_l^{(j)} + \hbar\kappa_{jl}^* a_j b_l^{(j)+}), \tag{2.3}$$

where $\psi_l^{(j)}$ is the frequency of the reservoir mode l coupled to the radiation mode j, κ_{jl} are coupling constants between the radiation and the reservoir described by boson annihilation and creation operators $b_l^{(j)}$ and $b_l^{(j)+}$ respectively (rotating terms considered e.g. in [1–3] are not taken into account here).

The usual procedure of the elimination of the reservoir variables in the Heisenberg equations for a_j [6, 1, 2] leads to the Heisenberg-Langevin equations

$$\dot{a}_1 = -(i\omega_1+\gamma_1/2)a_1 - ikg^* a_1^{+k-1} a_2 + L_1,$$
$$\dot{a}_2 = -(i\omega_2+\gamma_2/2)a_2 - iga_1^k + L_2, \tag{2.4}$$

where the Langevin forces L_j are determined by

$$L_j = -i\sum_l \kappa_{jl} b_l^{(j)} \exp(-i\psi_l^{(j)}t); \tag{2.5}$$

here $b_l^{(j)}$ are the initial values of the reservoir operators and γ_j are the damping constants. The forces (2.4) fulfill the following conditions

$$\langle L_j^+(t)L_j(t')\rangle = \gamma_j\langle n_{dj}\rangle\delta(t-t'), \quad \langle L_j(t)\rangle = 0, \tag{2.6}$$

where the average is taken over the reservoir variables $b_l^{(j)}$ which are assumed to be Gaussian, $\langle n_{dj}\rangle = \langle b_l^{(j)+}b_l^{(j)}\rangle$.

The equations (2.4) are solved in such a way that the Laplace transformation of (2.4) is taken and the Laplace transforms of non-linear terms $-ikg^* a_1^{+k-1}a_2$ and $-iga_1^k$ are modified by the integration by parts where (2.4) is used again in the corresponding terms. Continuing successively this procedure we are able to obtain a solution up to arbitrary powers of g [3]. Making use of two steps one finds [3]

$$a_1(t) = \exp(-i\omega_1 t - \gamma_1 t/2)\,[a_1 - ikg^* G_{11}(t) a_1^{+k-1} a_2$$
$$+ k|g|^2 G_{12}^{(1)}(t)\,[a_1^k, a_1^{+k-1}]a_2^+ a_2 - k|g|^2 G_{12}^{(2)}(t) a_1^{+k-1} a_1^k + \sum_l w_{1l} R_l^{(1)}(t)],$$

$$a_2(t) = \exp(-i\omega_2 t - \gamma_2 t/2)\,[a_2 - igG_{21}(t) a_1^k$$
$$-|g|^2 G_{22}^{(1)}(t)\,[a_1^k, a_1^{+k}]a_2 + \sum_l w_{2l} R_l^{(2)}(t)], \tag{2.7}$$

where a_j are the initial values of the corresponding operators, the identity

$$[a^l, a^{+k}] = k\sum_{j=0}^{l-1} a^j a^{+k-1} a^{l-1-j} = l\sum_{i=0}^{k-1} a^{+i} a^{l-1} a^{+k-1-i} \tag{2.8}$$

has been used and

$$G_{11}(t) = \frac{1}{\Gamma_{k-2,1}^{(1)}}(1-\exp(-\Gamma_{k-2,1}^{(1)}t)),$$

$$G_{12}^{(1)}(t) = \frac{1}{\Gamma_{k-2,1}^{(1)}}\left[\frac{1}{\Gamma_{2k-4,2}^{(1)}}(1-\exp(-\Gamma_{2k-4,2}^{(1)}t))\right.$$
$$\left.-\frac{1}{\Gamma_{k-2,1}^{(1)}}(\exp(-\Gamma_{k-2,1}^{(1)}t)-\exp(-\Gamma_{2k-4,2}^{(1)}t))\right],$$

$$G_{12}^{(2)}(t) = \frac{1}{\Gamma_{k-2,1}^{(1)}}\left[\frac{1}{\Gamma_{2k-2,0}^{(1)}}(1-\exp(-\Gamma_{2k-2,0}^{(1)}t))\right.$$

$$\left.-\frac{1}{\Gamma_{k,-1}^{(1)}}(\exp(-\Gamma_{k-2,1}^{(1)}t)-\exp(-\Gamma_{2k-2,0}^{(1)}t))\right],$$

$$G_{21}(t) = \frac{1}{\Gamma_{k,-1}^{(2)}}(1-\exp(-\Gamma_{k,-1}^{(2)}t)),$$

$$G_{22}^{(1)}(t) = \frac{1}{\Gamma_{k,-1}^{(2)}}\left[\frac{1}{\Gamma_{2k-2,0}^{(2)}}(1-\exp(-\Gamma_{2k-2,0}^{(2)}t))\right.$$

$$\left.-\frac{1}{\Gamma_{k-2,1}^{(2)}}(\exp(-\Gamma_{k,-1}^{(2)}t)-\exp(-\Gamma_{2k-2,0}^{(2)}t))\right],$$

$$R_l^{(j)}(t) = \frac{1}{i(\psi_l^{(j)}-\omega_j)-\gamma_j/2}(1-\exp[-i(\psi_l^{(j)}-\omega_j)+\gamma_j/2]t),$$

$$w_{jl} = -i\kappa_{jl}b_l^{(j)}, \tag{2.9}$$

where Γ are some compound damping constants, e.g. $\Gamma_{k-1,1}^{(1)} = (k-1)\gamma_1/2+\gamma_2/2$, $\Gamma_{k,0}^{(2)} = k\gamma_1/2$, $\Gamma_{k,-1}^{(2)} = (k\gamma_1-\gamma_2)/2$, etc. and the quantities $G_{lm}^{(j)}$ describe the damping of the j-th term in the m-th degree of decomposition for the l-th mode. The quantities $R_l^{(j)}$ describe the losses of the j-th mode. Those terms containing the commutators in (2.7) are typically quantum ones.

For the second order process we obtain by putting $k = 2$

$$a_1(t) = \exp(-i\omega_1 t-\gamma_1 t/2)\,[a_1-2ig^*G_{11}(t)a_1^+a_2$$

$$+4|g|^2G_{12}^{(1)}(t)a_1a_2^+a_2-2|g|^2G_{12}^{(2)}(t)a_1^+a_1^2+\sum_l w_{1l}R_l^{(1)}(t)],$$

$$a_2(t) = \exp(-i\omega_2 t-\gamma_2 t/2)\,[a_2-igG_{21}(t)a_1^2-2|g|^2G_{22}^{(1)}(t)$$

$$\times(2a_1^+a_1a_2+a_2)+\sum_l w_{2l}R_l^{(2)}(t)], \tag{2.10}$$

where now

$$G_{11}(t) = \frac{2}{\gamma_2}(1-\exp(-\gamma_2 t/2)),$$

$$G_{12}^{(1)}(t) = \frac{2}{\gamma_2^2}[1+\exp(-\gamma_2 t)-2\exp(-\gamma_2 t/2)],$$

$$G_{12}^{(2)}(t) = \frac{2}{\gamma_2}\left[\frac{1}{\gamma_1}(1-\exp(-\gamma_1 t))-\frac{1}{\gamma_1-\gamma_2/2}(\exp(-\gamma_2 t/2)-\exp(-\gamma_1 t))\right],$$

$$G_{21}(t) = \frac{1}{\gamma_1-\gamma_2/2}(1-\exp[-(\gamma_1-\gamma_2/2)t]),$$

$$G_{22}^{(1)}(t) = \frac{1}{\gamma_1-\gamma_2/2}\left[\frac{1}{\gamma_1}(1-\exp(-\gamma_1 t))-\frac{2}{\gamma_2}(\exp(-\gamma_1+\gamma_2/2)t-\exp(-\gamma_1 t))\right]. \tag{2.11}$$

Performing the decomposition of the functions G with respect to t up to t^2 ($G_{11}(t) = t-t^2[(k-2)\gamma_1/2+\gamma_2/2]/2$, $G_{12}^{(1)}(t) = G_{12}^{(2)}(t) = G_{22}^{(1)}(t) = t^2/2$, $G_{21}(t) = t-t^2(k\gamma_1-\gamma_2)/4$) and substituting $R_l^{(j)}(t) \approx$ t (i.e. $\psi_l^{(j)} \approx \omega_j$ is applied in (2.9); the term with t^2 cannot give any contribution to the characteristic function calculated up to t^2 since $\langle b_l^{(j)} \rangle = 0$), we obtain the power series solution [1, 2]

$$a_1(t) = \exp(-i\omega_1 t-\gamma_1 t/2)\Big\{a_1 - ikg^* t a_1^{+k-1} a_2 + L_1 t$$
$$+\frac{t^2}{2}[k|g|^2[a_1^k, a_1^{+k-1}]a_2^+ a_2 - k|g|^2 a_1^{+k-1} a_1^k + ikg^* \Gamma_{k-2,1}^{(1)} a_1^{+k-1} a_2]\Big\},$$

$$a_2(t) = \exp(-i\omega_2 t-\gamma_2 t/2)\Big\{a_2 - igta_1^k + L_2 t - \frac{t^2}{2}[|g|^2[a_1^k, a_1^{+k}]a_2 - ig\Gamma_{k,-1}^{(2)} a_1^k]\Big\},$$
(2.12)

where $\Gamma_{k-2,1}^{(1)}$ and $\Gamma_{k,-1}^{(2)}$ are given above and $L_j = L_j(0)$. Note that in [3] more general expressions containing the above mentioned rotating terms have been given.

Using (2.7) and the identity

$$\sum_l |\kappa_{jl}|^2 |R_l^{(j)}|^2 = \exp(\gamma_j t)-1, \tag{2.13}$$

which can be derived by substituting the integral over $\psi^{(j)}$ for the above sum and making use of the residuum theorem, we can prove that $[a_j(t), a_k^+(t)] = \delta_{jk}$, etc. (we have assumed $\gamma_1 \approx \gamma_2 \approx \gamma$ for simplicity). If the power series solution (2.12) is used for the calculation of the commutators, then the exponential function $\exp(-\gamma_j t)$ arising from the damping factors in (2.12) is only compensated by the quantity $\exp(\gamma_j t^2)$ arising from the terms $L_j t$ and it is $t^2 < t$ since $t \ll 1$. This is a consequence of the fact that in (2.12) the uniform reservoir spectrum ($\psi_l^{(j)} \approx \omega_j$) is assumed while in (2.7) the correct reservoir spectrum included in $R_l^{(j)}(t)$ is taken into account, i.e. it is correctly assumed that contributions of single reservoir oscillators to the resulting reservoir term are dependent on the frequency difference $\psi_l^{(j)} - \omega_j$.

The normal quantum characteristic function for separate modes,

$$C_{\mathcal{N}}(\beta_j, t) = \langle \exp(\beta_j a_j^+(t)) \exp(-\beta_j^* a_j(t)) \rangle, \tag{2.14}$$

(β_j is a complex parameter and the brackets mean the average over the initial complex amplitudes α_j, which are eigenvalues of a_j in the coherent state $|\alpha_j\rangle$, and over the reservoir variables), can be written, performing the average over the reservoir variables, decomposing the exponential functions and conserving terms up to the second order in β_j (strong field approximation) and using the Glauber-Sudarshan representation of the density matrix at $t = 0$, in the usual form [1–3]

$$C_{\mathcal{N}}(\beta_j, t) = \langle \exp[\beta_j \alpha_j^*(t) - \beta_j^* \alpha_j(t) - B_j(t)\,|\beta_j|^2 + C_j^*(t)\beta_j^2/2 + C_j(t)\beta_j^{*2}/2] \rangle, \tag{2.15a}$$

corresponding to the Glauber–Sudarshan quasi-distribution (through the Fourier transformation)

$$\phi_{\mathcal{N}}(\alpha_j, t) = (\pi K_j^{1/2}(t))^{-1} \left\langle \exp\left[-\frac{B_j(t)}{K_j(t)} |\alpha_j - \alpha_j(t)|^2 + \frac{C_j^*(t)(\alpha_j - \alpha_j(t))^2 + \text{c.c.}}{2K_j(t)} \right] \right\rangle; \tag{2.15b}$$

this describes the superposition of signal $\alpha_j(t)$ and noise $K_j(t) = B_j^2(t) - |C_j(t)|^2$ with correlated real and imaginary parts of the complex amplitude α_j and the brackets mean here the average over the initial complex amplitudes α_j. Here

$$\alpha_1(t) = \langle \alpha_1, \alpha_2 | \mathcal{N} a_1(t) | \alpha_1, \alpha_2 \rangle = \exp(-i\omega_1 t - \gamma_1 t/2)$$
$$\times \left\{ \alpha_1 - ikg^* t \alpha_1^{*k-1} \alpha_2 + \frac{t^2}{2} \left[|g|^2 \alpha_1 L_{k-1}^1(-|\alpha_1|^2) |\alpha_2|^2 - k|g|^2 \alpha_1^k \alpha_1^{*k-1}(1 + |\alpha_2|^2) + ikg^* \Gamma_{k-2,1}^{(1)} \alpha_1^{*k-1} \alpha_2 \right] \right\},$$

$$\alpha_2(t) = \langle \alpha_1, \alpha_2 | \mathcal{N} a_2(t) | \alpha_1, \alpha_2 \rangle = \exp(-i\omega_2 t - \gamma_2 t/2)$$
$$\times \left\{ \alpha_2 - igt\alpha_1^k - \frac{t^2}{2} \left[|g|^2 L_k^0(-|\alpha_1|^2)\alpha_2 - |g|^2 |\alpha_1|^{2k} \alpha_2 - ig\Gamma_{k,-1}^{(2)} \alpha_1^k \right] \right\},$$

$$B_1(t) = \langle \alpha_1, \alpha_2 | \mathcal{N} a_1^+(t) a_1(t) | \alpha_1, \alpha_2 \rangle = \exp(-\gamma_1 t)$$
$$\times \left\{ |\alpha_1|^2 - ikg^* t \alpha_1^{*k} \alpha_2 + ikgt\alpha_1^k \alpha_2^* + t^2 \left[k^2 |g|^2 L_{k-1}^0(-|\alpha_1|^2) \right.\right.$$
$$\times |\alpha_2|^2 + |g|^2 |\alpha_1|^2 L_{k-1}^1(-|\alpha_1|^2) |\alpha_2|^2 - k|g|^2 |\alpha_1|^{2k}(|\alpha_2|^2 + 1) + \frac{ikg^*}{2} \Gamma_{k-2,1}^{(1)} \alpha_1^{*k} \alpha_2$$
$$\left.\left. - \frac{ikg}{2} \Gamma_{k-2,1}^{(1)} \alpha_1^k \alpha_2^* \right] + \gamma_1 t^2 \langle n_{d1} \rangle \right\},$$

$$B_2(t) = \langle \alpha_1, \alpha_2 | \mathcal{N} a_2^+(t) a_2(t) | \alpha_1, \alpha_2 \rangle = \exp(-\gamma_2 t)(|g|^2 t^2 |\alpha_1|^{2k} + \gamma_2 t^2 \langle n_{d2} \rangle),$$

$$C_1(t) = \langle \alpha_1, \alpha_2 | \mathcal{N} a_1^2(t) | \alpha_1, \alpha_2 \rangle = \exp(-2i\omega_1 t - \gamma_1 t)$$
$$\times \left\{ \alpha_1^2 - 2ikg^* t \alpha_1^{*k-1} \alpha_1 \alpha_2 - ik(k-1) g^* t \alpha_1^{*k-2} \alpha_2 + \frac{t^2}{2} \left[\frac{|g|^2}{k+1} \alpha_1^2 L_{k-1}^2(-|\alpha_1|^2) |\alpha_2|^2 \right.\right.$$
$$- k(k-1)|g|^2 \alpha_1^{*k-2} \alpha_1^k |\alpha_2|^2 - 2k|g|^2 \alpha_1^{*k-1} \alpha_1^{k+1}(|\alpha_2|^2 + 1) - k(k-1)|g|^2 \alpha_1^{*k-2} \alpha_1^k$$
$$+ |g|^2 \alpha_1^2 L_{k-1}^1(-|\alpha_1|^2) |\alpha_2|^2 - 2k^2 g^{*2} \alpha_1^{*2k-2} \alpha_2^2$$
$$\left.\left. + 2ikg^* \Gamma_{k-2,1}^{(1)} \alpha_1^{*k-1} \alpha_1 \alpha_2 + ik(k-1) g^* \Gamma_{k-2,1}^{(1)} \alpha_1^{*k-2} \alpha_2 \right] \right\},$$

$$C_2(t) = \langle \alpha_1, \alpha_2 | \mathcal{N} a_2^2(t) | \alpha_1, \alpha_2 \rangle = -g^2 t^2 \alpha_1^{2k} \exp(-2i\omega_2 t - \gamma_2 t), \tag{2.16}$$

where $\langle n_{dj}\rangle$ is the mean number of reservoir oscillators coupled to the j-th radiation mode, $|\alpha_1, \alpha_2\rangle$ is the initial coherent state for the whole field composed of two modes, $\mathcal{N}$ denotes the normal ordering operation in the initial operators using the commutators and L_n^k are the Laguerre polynomials.

Using solutions (2.7) quite similarly, one obtains for instance

$$\alpha_2(t) = \exp(-i\omega_2 t - \gamma_2 t/2)\,\{\alpha_2 - igG_{21}(t)\alpha_1^k - |g|^2 G_{22}^{(1)}(t)\,[L_k^0(-|\alpha_1|^2)\alpha_2 - |\alpha_1|^{2k}\alpha_2]\},$$
$$B_2(t) = \exp(-\gamma_2 t)\,|g|^2 G_{21}^2(t)\,|\alpha_1|^{2k} + (1-\exp(-\gamma_2 t))\,\langle n_{d2}\rangle,$$
$$C_2(t) = -g^2 G_{21}^2(t)\alpha_1^{2k}\exp(-2i\omega_2 t - \gamma_2 t), \tag{2.17}$$

etc.

For $k = 2$ we particularly have

$$a_1(t) = \exp(-i\omega_1 t - \gamma_1 t/2)\left\{a_1 - 2ig^* t a_1^+ a_2 + L_1 t + |g|^2 t^2 (2a_1 a_2^+ a_2 - a_1^+ a_1^2) + \frac{i}{2} g^* \gamma_2 t^2 a_1^+ a_2\right\},$$

$$a_2(t) = \exp(-i\omega_2 t - \gamma_2 t/2)\left\{a_2 - igta_1^2 + L_2 t - |g|^2 t^2 \times (2a_1^+ a_1 a_2 + a_2) + \frac{i}{2} g(\gamma_1 - \gamma_2/2) t^2 a_1^2\right\},$$

$$\alpha_1(t) = \exp(-i\omega_1 t - \gamma_1 t/2)\left\{\alpha_1 - 2ig^* t\alpha_1^*\alpha_2 + |g|^2 t^2 (2\alpha_1|\alpha_2|^2 - \alpha_1^*\alpha_1^2) + \frac{i}{2} g^*\gamma_2 t^2 \alpha_1^*\alpha_2\right\},$$

$$\alpha_2(t) = \exp(-i\omega_2 t - \gamma_2 t/2)\left\{\alpha_2 - igt\alpha_1^2 - |g|^2 t^2 (2|\alpha_1|^2\alpha_2 + \alpha_2) + \frac{i}{2} g(\gamma_1 - \gamma_2/2) t^2 \alpha_1^2\right\},$$

$$B_1(t) = \exp(-\gamma_1 t)\left[|\alpha_1|^2 + 2it(g\alpha_1^2\alpha_2^* - g^*\alpha_1^{*2}\alpha_2) + 2|g|^2 t^2 (4|\alpha_1|^2|\alpha_2|^2 - |\alpha_1|^4 + 2|\alpha_2|^2) + \frac{i}{2}\gamma_2 t^2 (g^*\alpha_1^{*2}\alpha_2 - g\alpha_1^2\alpha_2^*) + \gamma_1 t^2 \langle n_{d1}\rangle\right],$$

$$B_2(t) = \exp(-\gamma_2 t)\,(|g|^2 t^2 |\alpha_1|^4 + \gamma_2 t^2 \langle n_{d2}\rangle),$$

$$C_1(t) = \exp(-2i\omega_1 t - \gamma_1 t)\left\{\alpha_1^2 - 4ig^* t|\alpha_1|^2\alpha_2 - 2ig^* t\alpha_2 - 4g^{*2} t^2 \alpha_1^{*2}\alpha_2^2 + |g|^2 t^2 (4\alpha_1^2|\alpha_2|^2 - 2|\alpha_1|^2\alpha_1^2 - \alpha_1^2) + ig^*\gamma_2 t^2 |\alpha_1|^2\alpha_2 + ig^*\frac{\gamma_2}{2} t^2 \alpha_2\right\},$$

$$C_2(t) = -g^2 t^2 \alpha_1^4 \exp(-2i\omega_2 t - \gamma_2 t). \tag{2.18}$$

Expressions (2.16) and (2.18) have been used in [1–3] to discuss the statistical properties of the k-th harmonic putting $\alpha_2 = 0$ (there are no k-th harmonic photons at $t = 0$). For incident coherent light (the brackets are omitted in (2.15a, b) in this case) we observe that both the modes are generally getting the quantum noise proportionally to the incident intensity with further addition of the reservoir noise. But neglecting the vacuum contribution in mode 2, i.e. putting $a_2 = 0$ in (2.7), (2.10) and (2.12) we see that $a_2(t)$ depends on the annihilation operator a_1 only while $a_1(t)$ depends also on the creation operator a_1^+. Thus in this case, neglecting the reservoir contribution supporting generally the noise component, the characteristic function (2.14) for mode 2 is already in the normal order in the initial operators and mode 2 is in the coherent state $|\alpha_2(t)\rangle$ and it shows a tendency to be coherent. Mode 1, however, corresponding to the pumping radiation is losing coherence proportionally to its intensity.

For the k-th subharmonic we put $\alpha_1 = 0$ (there are no subharmonic photons at $t = 0$) and we have from (2.16)

$$B_1(t) = \exp(-\gamma_1 t)\,(kk!|g|^2t^2|\alpha_2|^2+\gamma_1 t^2\langle n_{d1}\rangle),$$

$$B_2(t) = \exp(-\gamma_2 t)\gamma_2 t^2\langle n_{d2}\rangle,$$

$$C_1(t) = -\exp(-2i\omega_1-\gamma_1 t)ik(k-1)g^*\left(t-\frac{t^2}{2}\Gamma^{(1)}_{k-2,1}\right)\alpha_2\delta_{k,2},$$

$$C_2(t) = 0, \tag{2.19a}$$

where $\delta_{k,2}$ is the Kronecker delta. Thus neglecting the reservoir contribution we observe that the pumping radiation (mode 2) has a tendency to be coherent while the k-th subharmonic (mode 1) is getting the quantum noise in the process of interaction similarly as in the case of parametric processes [1, 2]. We also observe a qualitative difference in behaviour of the subharmonics mode for $k = 2$ and $k > 2$ where $C_1(t)$ is missing. Similar qualitative differences have been also found in [4].

Based on the non-power solution we similarly obtain

$$B_1(t) = kk!|g|^2G_{11}^2(t)\,|\alpha_2|^2\exp(-\gamma_1 t)+(1-\exp(-\gamma_1 t))\,\langle n_{d1}\rangle,$$

$$B_2(t) = (1-\exp(-\gamma_2 t))\,\langle n_{d2}\rangle,$$

$$C_1(t) = -ik(k-1)g^*G_{11}(t)\alpha_2\delta_{k,2}\exp(-2i\omega_1 t-\gamma_1 t),$$

$$C_2(t) = 0. \tag{2.19b}$$

3. *Schrödinger (generalized Fokker–Planck equation) approach*

The generalized Fokker–Planck equation for the Glauber–Sudarshan quasi-distribution related to normal ordering of field operators for the process under consideration can be written in the usual way [6] in the following form

$$\frac{\partial\phi_{\mathcal{N}}}{\partial t} = \left\{\left[\frac{\partial}{\partial\alpha_1}(\gamma_1/2+i\omega_1)\alpha_1+\frac{\partial}{\partial\alpha_2}(\gamma_2/2+i\omega_2)\alpha_2\right]+\text{c.c.}\right.$$

$$+\gamma_1\langle n_{d1}\rangle \frac{\partial^2}{\partial\alpha_1\partial\alpha_1^*} +\gamma_2\langle n_{d2}\rangle \frac{\partial^2}{\partial\alpha_2\partial\alpha_2^*} + \Bigg[ig\alpha_1^k \frac{\partial}{\partial\alpha_2}$$

$$-ig^* \sum_{j=1}^{k} (-1)^j \binom{k}{j} \alpha_1^{*k-j}\alpha_2 \frac{\partial^2}{\partial\alpha_1^j}\Bigg] +\text{c.c.}\Bigg\}\, \phi_{\mathcal{N}}, \qquad (3.1)$$

where the first four terms correspond to the equations of motion for two damped harmonic oscillators, the fifth and sixth terms represent the reservoir contributions and the remaining terms characterize the optical non-linear process and they are obtained from the equation of motion for the density matrix ϱ,

$$i\hbar \frac{\partial\varrho}{\partial t} = [H, \varrho], \qquad (3.2)$$

if

$$\frac{1}{i\hbar}[\hbar g a_1^k a_2^+ + \hbar g^* a_1^{+k} a_2, \varrho] \qquad (3.3)$$

is transformed to the antinormal form and q-c-number correspondence of the coherent state technique is applied ($\phi_{\mathcal{N}}(\alpha) = \varrho^{(\mathscr{A})}(a \to \alpha, a^+ \to \alpha^*)/\pi$) [6, 7], i.e. one applies the identities

$$\varrho^{(\mathscr{A})} a_1^k = \mathscr{A}\left\{\left(a_1 - \frac{\partial}{\partial a_1^+}\right)^k \varrho\right\},$$

$$a_1^{+k}\varrho^{(\mathscr{A})} = \mathscr{A}\left\{\left(a_1^+ - \frac{\partial}{\partial a_1}\right)^k \varrho\right\},$$

$$[a_2, \varrho] = \frac{\partial\varrho}{\partial a_2^+}, \quad [\varrho, a_2^+] = \frac{\partial\varrho}{\partial a_2}, \qquad (3.4)$$

where $\mathscr{A}$ denotes the antinormal ordering operation. It can be shown that the quasi-distribution $\phi_{\mathcal{N}}$ exists in this case for the approximations used in [1 — 3]. In the Markoff approximation (3.1) can be written in the following form

$$\frac{\partial\phi_{\mathcal{N}}}{\partial t} = \Bigg\{\left[\frac{\partial}{\partial\alpha_1}(\gamma_1/2+i\omega_1)\alpha_1 + \frac{\partial}{\partial\alpha_2}(\gamma_2/2+i\omega_2)\alpha_2\right] +\text{c.c.}$$

$$+\gamma_1\langle n_{d1}\rangle \frac{\partial^2}{\partial\alpha_1\partial\alpha_1^*} +\gamma_2\langle n_{d2}\rangle \frac{\partial^2}{\partial\alpha_2\partial\alpha_2^*} + \Bigg[ig\alpha_1^k \frac{\partial}{\partial\alpha_2}$$

$$+ikg^*\alpha_1^{*k-1}\alpha_2 \frac{\partial}{\partial\alpha_1} - \frac{i}{2} g^* k(k-1)\alpha_1^{*k-2}\alpha_2 \frac{\partial^2}{\partial\alpha_1^2}\Bigg] +\text{c.c.}\Bigg\}\, \phi_{\mathcal{N}}. \qquad (3.5)$$

Particularly for $k = 2$ we obtain

$$\frac{\partial \phi_{\mathcal{N}}}{\partial t} = \left\{ \left[\frac{\partial}{\partial \alpha_1} [(\gamma_1/2 + i\omega_1)\alpha_1 + 2ig^*\alpha_1^*\alpha_2] + \frac{\partial}{\partial \alpha_2} [(\gamma_2/2 + i\omega_2)\alpha_2 + ig\alpha_1^2] \right] \right.$$

$$\left. + \text{c.c.} + \gamma_1 \langle n_{d1} \rangle \frac{\partial^2}{\partial \alpha_1 \partial \alpha_1^*} + \gamma_2 \langle n_{d2} \rangle \frac{\partial^2}{\partial \alpha_2 \partial \alpha_2^*} - ig^*\alpha_2 \frac{\partial^2}{\partial \alpha_1^2} + \text{c.c.} \right\} \phi_{\mathcal{N}} \quad (3.6)$$

and performing the Fourier transformation,

$$\phi_{\mathcal{N}}(\alpha_1, \alpha_2, t) = \frac{1}{(\pi^2)^2} \int C_{\mathcal{N}}(\beta_1, \beta_2, t) \prod_{j=1}^{2} \exp(\beta_j^*\alpha_j - \beta_j\alpha_j^*) d^2\beta_j, \quad (3.7)$$

we have the equation for the normal characteristic function

$$\frac{\partial C_{\mathcal{N}}}{\partial t} = \left\{ \left[(-\gamma_1/2 + i\omega_1)\beta_1 \frac{\partial}{\partial \beta_1} + (-\gamma_2/2 + i\omega_2)\beta_2 \frac{\partial}{\partial \beta_2} \right] \right.$$

$$+ \text{c.c.} - \gamma_1 \langle n_{d1} \rangle |\beta_1|^2 - \gamma_2 \langle n_{d2} \rangle |\beta_2|^2 + \left[ig\beta_1^2 \frac{\partial}{\partial \beta_2} \right.$$

$$\left. \left. + ig^*\beta_2 \frac{\partial^2}{\partial \beta_1^2} - 2ig^*\beta_1^* \frac{\partial^2}{\partial \beta_1 \partial \beta_2^*} \right] - \text{c.c.} \right\} C_{\mathcal{N}}. \quad (3.8)$$

This equation has been solved by means of iterations starting from the solution with $g = 0$, i.e. from

$$C_{\mathcal{N}}^{(0)}(\beta_1, \beta_2. t) = \prod_{j=1}^{2} \exp[-\langle n_{dj} \rangle (1 - e^{-\gamma_j t}) |\beta_j|^2 + \beta_j \alpha_{j0}^*(t) - \beta_j^* \alpha_{j0}(t)],$$

$$\alpha_{j0}(t) = \alpha_j \exp(-i\omega_j t - \gamma_j t/2). \quad (3.9)$$

The first iteration provides using moreover the method of characteristics together with the assumption of small t

$$C_{\mathcal{N}}^{(1)}(\beta_1, \beta_2, t) = \exp\{-\langle n_{d1} \rangle (1 - e^{-\gamma_1 t}) |\beta_1|^2 - \langle n_{d2} \rangle (1 - e^{-\gamma_2 t}) |\beta_2|^2$$

$$+ [\beta_1 \alpha_1^* \exp(-\gamma_1 t/2 + i\omega_1 t) + 2igt\beta_1\alpha_1\alpha_2^* \exp(-\gamma_1 t/2 + i\omega_1 t)$$

$$+ \beta_2\alpha_2^* \exp(-\gamma_2 t/2 + i\omega_2 t) + ig^*t\beta_2\alpha_1^{*2} \exp(-\gamma_2 t/2 + i\omega_2 t)] - \text{c.c.}$$

$$+ igt\beta_1^2\alpha_2^* \exp(-\gamma_1 t + 2i\omega_1 t) + \text{c.c.}\}. \quad (3.10)$$

It should be noted that no strong intensity assumption is needed here in contrast to the Heisenberg–Langevin method.

Thus

$$\alpha_1(t) = \exp(-i\omega_1 t - \gamma_1 t/2)(\alpha_1 - 2ig^* t\alpha_1^* \alpha_2),$$

$$\alpha_2(t) = \exp(-i\omega_2 t - \gamma_2 t/2)(\alpha_2 - igt\alpha_1^2),$$

$$B_1(t) = \langle n_{d1}\rangle(1 - e^{-\gamma_1 t}),$$

$$B_2(t) = \langle n_{d2}\rangle(1 - e^{-\gamma_2 t}),$$

$$C_1(t) = -2ig^* t\alpha_2 \exp(-2i\omega_1 t - \gamma_1 t),$$

$$C_2(t) = 0. \tag{3.11}$$

Comparing this with (2.18) we find that we have the identical evolution of the complex amplitudes $\alpha_j(t)$ up to the first order in gt. Further we observe that both the modes are statistically independent in this case. For B_j and C_j (3.11) provides more compact solutions without using the strong intensity approximation for calculation of the characteristic function so the reservoir is described more fully. We also see that up to gt operators $a_1(t)$ and $a_2(t)$ for $a_2 = 0$ (second harmonic generation case without the vacuum contribution) are dependent only on annihilation operators and consequently B_j depend on reservoir properties in the above way only and $C_j = 0$. For $\alpha_1 = 0$ (second subharmonic generation case) the expressions for B_j and C_j given in (2.19a) agree with those in (3.11), there is only a difference in the reservoir contributions as discussed above. Similar results have been obtained making the second iteration and they are in agreement with the results of Section 2. Detailed discussion of the statistical properties of higher harmonics and subharmonics based on a more compact solution of the Fokker–Planck equation is in preparation.

So we have the same conclusion that the pumping wave has a tendency to be coherent (neglecting the reservoir contribution) and the second subharmonic is getting the physical vacuum noise.

The question of the determination of the photocounting statistics corresponding to the quasi-distribution of the form (2.15b) has been discussed in [8, 9].

4. *Realistic description of laser light*

The above given description is rather idealized since it assumes two modes for the incident and outgoing radiation. In order to describe the spectral structure of radiation we may use the phase diffusion model [10, 11].

We consider ω as a fluctuating quantity and substitute $\omega t \to \omega t + \int_0^t \Delta\omega(t')dt' = \omega t + \varphi(t)$, where $\varphi(t) = \int_0^t \Delta\omega(t')\,dt'$ is a fluctuating phase related to the fluctuating part $\Delta\omega$ of the frequency. Assuming that $\Delta\omega(t) = \sum_n c_n \varphi_n(t)$ ($\varphi_n(t)$ being an orthonormal set of real functions and c_n Gaussian real random variables) is a Gaussian process, it is well-known that ($\Delta\omega(0) = 0$ is assumed)

$$\langle \exp(-i\int_0^t \Delta\omega(t')dt')\rangle = \langle\prod_n \exp(-ic_n\lambda_n)\rangle$$

$$= \prod_n \int_{-\infty}^{+\infty} (\sqrt{2\pi}\,\sigma_n)^{-1} \exp\left(-\frac{c_n^2}{2\sigma_n^2} - i\lambda_n c_n\right) dc_n = \prod_n \exp(-\tfrac{1}{2}\sigma_n^2\lambda_n^2)$$

$$= \exp(-\tfrac{1}{2}\iint_0^t \sum_n \sigma_n^2\varphi_n(t')\varphi_n(t'')dt'dt'') = \exp(-\tfrac{1}{2}\iint_0^t \langle\Delta\omega(t')\Delta\omega(t'')\rangle dt'dt''), \quad (4.1)$$

where $\lambda_n = \int_0^t \varphi_n(t')dt'$, σ_n is the standard deviation of c_n and $\sigma_n^2 = \langle c_n^2\rangle$. Further assuming the Markoffian property of the process, $\langle\Delta\omega(t')\Delta\omega(t'')\rangle = 2D\delta(t'-t'')$, D being the diffusion constant of the phase fluctuations equal to the spectral halfwidth Γ, we have for the radiation spectrum of a damped wave $\sqrt{I_0}\exp(-i\omega_0 t - i\varphi(t) - \gamma|t|/2)$ with the diffusing phase $\varphi(t)$

$$G(\omega) = \int_{-\infty}^{+\infty} \langle a^+(t)a(0)\rangle \exp(-i\omega t)dt = I_0 \int_{-\infty}^{+\infty} \exp[i(\omega_0-\omega)t$$

$$-\gamma|t|/2 - \Gamma|t|]dt = \frac{2\Gamma+\gamma}{(\Gamma+\gamma/2)^2+(\omega-\omega_0)^2}, \quad (4.2)$$

which is the Lorentzian spectrum. If the reservoir damping constant is negligible compared to 2Γ, we see from(4.1) and (2.2) ($\omega_2 = k\omega_1$, $\Delta\omega_2 = k\Delta\omega_1$) that the spectral halfwidth of the k-th harmonic is $\Gamma_2 = k^2\Gamma_1$ (Γ_1 being the halfwidth of the pumping radiation), i.e.

$$G_1(\omega) = \frac{2\Gamma_1}{\Gamma_1^2+(\omega-\omega_1)^2}, \quad G_2(\omega) = \frac{2(k^2\Gamma_1)}{(k^2\Gamma_1)^2+(\omega-k\omega_1)^2} \quad (4.3)$$

as has been shown for $k = 2$ calculating the Glauber–Sudarshan quasi-probability function in [5]. Thus the second-order spectrum of k-th harmonic is k^2 broader than that of the pumping radiation. Similarly the spectrum of the k-th subharmonic is k^2 narrower than that of the pumping radiation. In the first case we have an example of a non-stationary field having a tendency to be coherent with an arbitrary spectral composition in the second case we have an example of the field with increasing level of noise and improving second-order temporal coherence (a chaotic field can be fully coherent in the second order but not in higher orders).

Finally we note that higher-order terms with respect to t in (2.12) give spectral corrections proportional to derivatives of (4.2) with respect to ω, while solutions (2.7) lead to a sum of Lorentzian spectra.

Note added in proof: Results obtained in Section 2 for mode 1 are related to the used method of calculation of the characteristic function assuming rather strong field. This excludes the so-called anticorrelation effect which has been found recently [12, 13] and which can also follow from results of Section 3 if $\gamma_j = \langle n_{dj} \rangle = 0$ (from (3.11) it holds that $\langle a_1^{+2}(t) a_1^2(t) \rangle - \langle a_1^+(t) a_1(t) \rangle^2 = -2\, ig^* t \alpha_1^{*2} \alpha_2 + 2\, igt\, \alpha_1^2 \alpha_2^*$ up to gt). In this case $\varphi_{\mathcal{N}}$ does not exist for mode 1 and quasi-distribution $\varphi_{\mathcal{A}}$ related to the antinormal ordering should be adopted, $B_1 \to B_1 + 1$.

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