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Quantum superposition states in third-harmonic generation

S. T. Gevorkyan
Institute for Physical Research, National Academy of Sciences of Armenia, Ashtarak-2 378410, Armenia
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We investigate numerically the distribution functions of photon number and phase in third-harmonic generation in the region of instability. It is shown that in this system the quantum superposition states for the third harmonic and fundamental mode can appear. [S1050-2947(98)00910-X]

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I. INTRODUCTION

The production and detection of nonclassical light continues to be an important topic in quantum optics. The quantum-mechanical superposition states [1] are nonclassical states of light which are of great interest currently.

Wolinsky and Carmichael have shown that the quantum-mechanical superposition states can be obtained during the generation of a second subharmonic in the case of a large nonlinearity [2]. The second subharmonic has two stable stationary classical solutions above the threshold of generation for the radiation field amplitude. In the region of strong quantum noises (large nonlinearities of interaction) the system is in a coherent state of superposition of two components. In the weak noise region (small nonlinearities) the superposition state goes to a state which is a classical mixture of these components.

The possibility to obtain the superposition states in the process of two-photon absorption for the coherent parametric excitation case was shown by Knight and co-workers [3]. In this paper the dynamics of the Wigner function for this process was investigated. It was shown that the competitive one-photon absorption process even if it is weak destroys with time the superposition state.

The quantum-mechanical superposition states were investigated also in Refs. [4–12]. Recently superposition states were observed experimentally with a single laser-cooled trapped ion [13] and by excitation of an atomic electron to give a coherent superposition of Rydberg states [14].

In Ref. [15] the dynamics of distribution of the phases of interacting modes for the process of the second-harmonic generation was investigated in the positive P representation [16]. The second harmonic, in contrast to the second subharmonic, has one classical solution for the field amplitude which becomes unstable above the bifurcation point of the optical system [17,18]. The system has in this domain only quantum solutions and is a macroscopic quantum object. For the fundamental mode and second harmonic a superposition state will be realized at large times. As distinct from the process of the second subharmonic, the quantum superposition state can be obtained in the case of a small nonlinearity.

In Ref. [19], Mlynek and co-workers used the method of quantum trajectories [20] to investigate the Wigner function for the third subharmonic. The classical treatment gives for the third subharmonic four steady solutions (one of which is the vacuum state). By demonstration of the quantum trajectories of an optical system the authors have shown that in the system a classical mixture of the states is realized, i.e., the system spends most of its time localized close to one of the classical solutions.

In the present paper the dynamics of fluctuations of the photon number and phases is investigated for the process of third-harmonic generation (THG). With this purpose we simulate in the positive P representation the Langevin equations of an optical system. This method was proposed by Dörfle and Schenzle to calculate the dynamics of the mean number of photons and quadrature amplitude of the field in the second-harmonic generation [21]. Various methods of calculations are given in Refs. [22–26]. In the present work we show that the fundamental mode, and the third harmonic, above the bifurcation point of an optical system at large times are in a quantum-mechanical superposition state. We investigate also joint fluctuations of phases of interacting modes.

II. BASIC EQUATIONS

We consider a double resonant third-harmonic generation in which three photons of frequency ω_1 in the fundamental mode a_1 can annihilate to produce a photon with the frequency $\omega_2=3\omega_1$ in the third-harmonic mode a_2. The fundamental mode is resonantly driven by an external classical field. The interaction of the fundamental mode with that of the third harmonic in a nonlinear $\chi^{(3)}$ medium is described by the following Hamiltonian:

$$H=\hbar\omega_1 a_1^\dagger a_1+3\hbar\omega_1 a_2^\dagger a_2+H_{\rm int}+H_{\rm loss},$$

$$H_{\rm int}=i\hbar\chi(a_1^3 a_2^\dagger-a_1^{\dagger 3}a_2)+i\hbar(Ee^{-i\omega_1 t}a_1^\dagger-E^*e^{i\omega_1 t}a_1),$$

$$H_{\rm loss}=a_1\Gamma_1^\dagger+a_1^\dagger\Gamma_1+a_2\Gamma_2^\dagger+a_2^\dagger\Gamma_2, \qquad (1)$$

were χ is the resulting coupling constant proportional to the third-order susceptibility $\chi^{(3)}$, E is the amplitude of the driving field at the frequency ω_1, and Γ_i, $\Gamma_i^\dagger$ ($i=1,2$) are reservoir operators for the fundamental and third-harmonic modes, which will give rise to the cavity damping constants γ_1 and γ_2, respectively.

Using standard techniques for the Hamiltonian (1), we obtain the master equation for the density matrix of the system. Furthermore, in the positive P representation, this equation is converted to a Fokker-Planck equation for the quasiprobability distribution function $P(\alpha_1,\alpha_2,\beta_1,\beta_2,t)$, where α_i and β_i ($i=1,2$) are the independent complex variables corresponding to a_i and $a_i^\dagger$, respectively. Using the Ito rules, from the Fokker-Planck equation, we can obtain the Langevin stochastic equations for the α_i and β_i variables as [27]

$$\begin{aligned} d\alpha_1 &= (\varepsilon-\alpha_1-3k\beta_1^2\alpha_2)d\tau+\sqrt{-6k\beta_1\alpha_2}\xi_1(\tau)\sqrt{d\tau} \\ &\quad+\sqrt{-6k\alpha_2}\eta_1(\tau)\sqrt[3]{d\tau}, \\ d\beta_1 &= (\varepsilon^*-\beta_1-3k\alpha_1^2\beta_2)d\tau+\sqrt{-6k\alpha_1\beta_2}\xi_2(\tau)\sqrt{d\tau} \\ &\quad+\sqrt[3]{-6k\beta_2}\eta_2(\tau)\sqrt[3]{d\tau}, \\ d\alpha_2 &= (-r\alpha_2+k\alpha_1^3)d\tau, \\ d\beta_2 &= (-r\beta_2+k\beta_1^3)d\tau. \end{aligned} \tag{2}$$

The variable $\tau=\gamma_1 t$ is the scaled time, $\varepsilon=E/\gamma_1$, $k=\chi/\gamma_1$, $r=\gamma_2/\gamma_1$, and $\xi_1(\tau)$, $\xi_2(\tau)$, $\eta_1(\tau)$, $\eta_2(\tau)$ are the independent Langevin sources of the noise with the following nonzero correlation functions:

$$\langle\xi_1^2\rangle=\langle\xi_2^2\rangle=1, \quad \langle\eta_1^3\rangle=\langle\eta_2^3\rangle=1. \tag{3}$$

The system of equations (2) without the noise terms has for large times stable stationary solutions for the photon numbers and phases,

$$n_j=\alpha_j\beta_j, \quad \phi_j=-\ln(\alpha_j/\beta_j)/2i, \tag{4}$$

only in the case of weak perturbation fields $\varepsilon<\varepsilon_{\rm cr}$, where $\varepsilon_{\rm cr}$ is the critical value of the perturbation (the Hopf's point of bifurcation) determined by the following formula:

$$\varepsilon_{\rm cr}=\sqrt[4]{r/6k^2}[(1+r)^{1/4}+\tfrac{1}{2}(1+r)^{5/4}]. \tag{5}$$

In the case of a strong perturbation $\varepsilon>\varepsilon_{\rm cr}$, small fluctuations of the phases of the fundamental and the third harmonic do not damp in time [the system loses stability near the stationary solutions of Eqs. (2) without the noise terms], while the semiclassical solutions for the numbers of photons turn into the auto-oscillation regime [27].

III. DYNAMICS OF FLUCTUATIONS AND FORMATION OF QUANTUM-MECHANICAL SUPERPOSITION STATE

We proceed with the calculation of the dynamics of the distribution function of the fundamental mode phase. We calculate this function with the help of the following obvious formula:

$$P(\phi_1,\tau)=\lim_{\substack{N\to\infty\\ \Delta\phi_1\to 0}}\left(\frac{S_N(\tau)}{N}\right). \tag{6}$$

Here $P(\phi_1,\tau)$ is the density of phase distribution at the moment of time τ, $S_N(\tau)$ is the number of those realizations of Eqs. (2) which at the moment of time τ are in the phase element $\Delta\phi_1$ with the point ϕ_1 inside. Figure 1 shows the dynamics of the distribution function of the fundamental mode phase for the Gaussian initial condition of both modes:

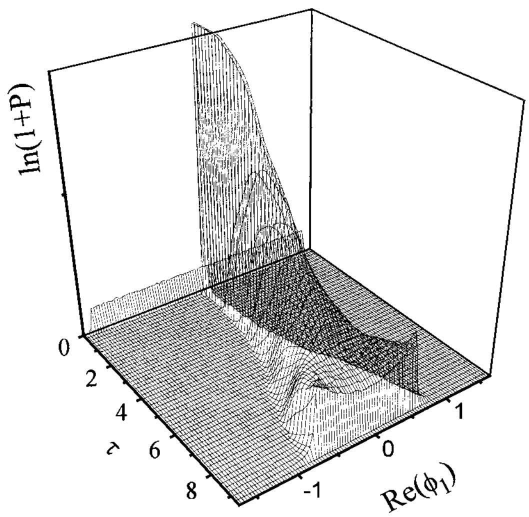

FIG. 1. Dynamics of distribution function of phase for the fundamental mode in the case of an initial Gaussian state of both modes and for following values of parameters: $r=1$, $k^2=10^{-9}$, $\varepsilon=380$. The function is calculated with the help of 100 000 independent trajectories of Eqs. (2).

$$\langle\alpha_i(0)\rangle=\langle\beta_i(0)\rangle=0, \quad \langle\beta_i(0)\alpha_j(0)\rangle=2\delta_{ij} \quad (i,j=1,2). \tag{7}$$

Here and below the following values of parameters are used for calculations: $r=1$, $k^2=10^{-9}$, $\varepsilon=380$. At these values the critical perturbation is $\varepsilon_{\rm cr}\approx 270$.

At $\tau=0$ we have a uniform distribution of phases in the interval $(-\pi/2,\pi/2)$. After some very small period the fundamental mode moves, due to the classical perturbation field, towards the coherent state with very narrow phase distribution. Then the system gradually moves towards the state with the two most probable values of the phase. States with two components are formed in the system. Hereafter the distribution function does not vary.

An incoherent superposition (classical mixture) is the limiting case of quantum superposition states where the time of the localization of the system in component states exceeds greatly the time of transition between them [19]. Since during the transition it is impossible to determine in which component the system is, the components interfere in the case where the transition time is of the order of the localization time. The system becomes a quantum object.

In Fig. 2 the dynamics of a certain realization of the fundamental mode phase is shown. The dashed lines correspond to the two most probable values of the phase. Time of transition of the system between these states is about the stay time in them. This indicates that in the system a coherent superposition of these states is realized.

The fact that a classical mixture is localized in its components is a consequence of the damping of fluctuations in an optical system. When a damping arises in a quantum superposition state, the interference between the components is destroyed and the system decays into the classical mixture of the components [3]. For the process of THG in the instability

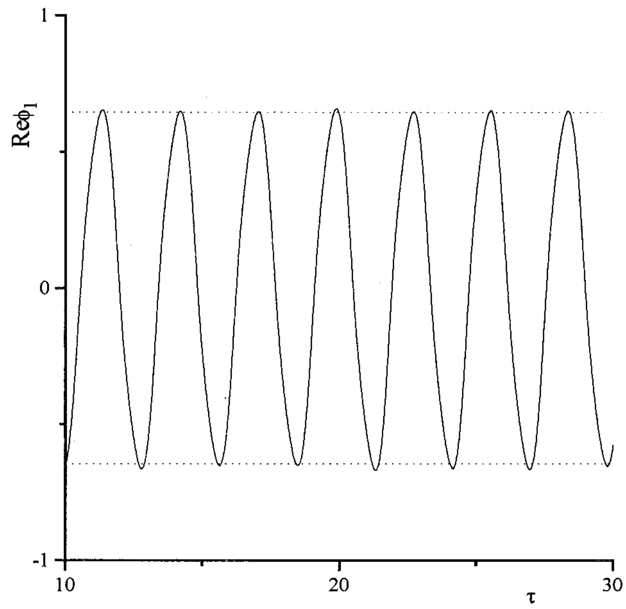

FIG. 2. Realization of a phase for the fundamental mode. The dashed lines lead to the two most probable values of the phase. The function is calculated for the case of parameters of Fig. 1.

region, small fluctuations of phases of interacting modes do not damp [27] which means quantum-mechanical interference between state components of the modes exists.

In Fig. 3 [curve (1)] the density of phase distribution of the fundamental mode is shown at the moment of time $\tau = 9.5$. It is approximated by the following function:

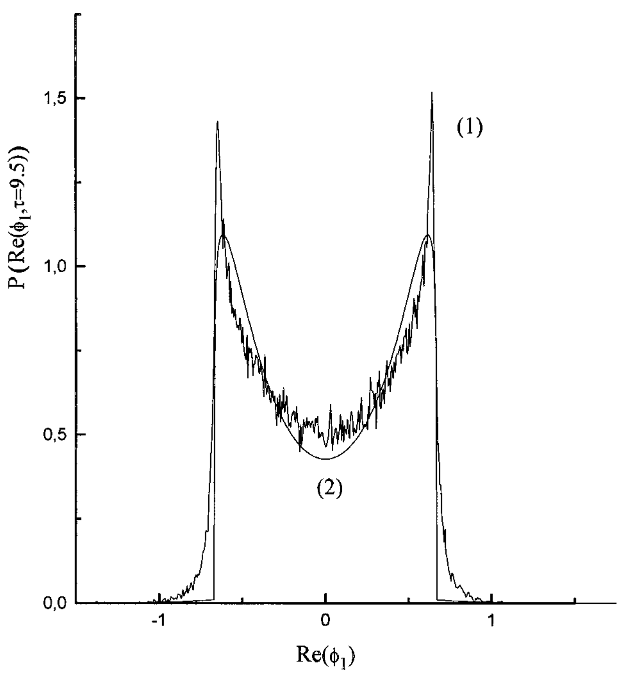

FIG. 3. Phase distribution function for the fundamental mode at the moment of time $\tau = 9.5$ (curve 1) and for values of parameters of Fig. 1. 100 000 trajectories of Eqs. (2). Curve (2) represents approximation of this function with the help of formula (8). Accuracy of the approximation is 7%.

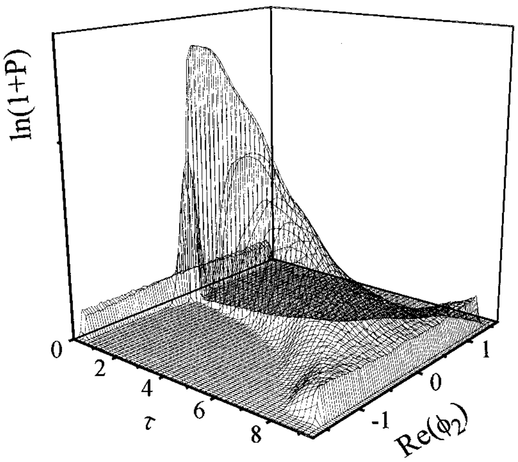

FIG. 4. Dynamics of phase distribution for the third harmonic in the case of an initial Gaussian state of both modes and for values of parameters of Fig. 1. 100 000 trajectories.

$$P(\mathrm{Re}\ \phi_1) = N\{F^2(\phi_0 + \mathrm{Re}\ \phi_1) + F^2(\phi_0 - \mathrm{Re}\ \phi_1) + 2\cos(\psi) F(\phi_0 + \mathrm{Re}\ \phi_1) F(\phi_0 - \mathrm{Re}\ \phi_1)\}. \tag{8}$$

Here the first two terms determine the components of the system state with a positive or negative value of the phase, the third term is determining the interference between these components, N is the normalization factor, $2\cos(\psi)$ is the factor of interference between the two states, and

$$F(\phi) = \begin{cases} \dfrac{b^a}{\Gamma(a)}\ \phi^{a-1} e^{-b\phi} & \text{at } \phi > 0 \\ 0 & \text{at } \phi \leq 0 \end{cases} \tag{9}$$

is the density of Γ distribution [28]. In the approximation used the above quantities are equal to $\phi_0 \approx 0.67$, $2\cos(\psi) \approx 1.57$, $a \approx 1.10$, $b \approx 2.03$. In this case a strong superposition state $2\cos(\psi) > 1$ will be realized in the system.

Dynamics of the phase distribution of the third harmonic is shown in Fig. 4. Here, as well as in the case of the fundamental mode, the quantum-mechanical superposition state will be formed in the system at large times. At the moment of time $\tau = 9.5$ the quantities of approximation of the phase distribution function of the third harmonic entering the formula (8) are equal to $\phi_0 \approx 1.51$, $2\cos(\psi) \approx 0.11$, $a \approx 1.42$, $b \approx 1.18$. In this case a strong superposition state $2\cos(\psi) < 1$ will be realized in the system.

The densities of the joint distribution of photon numbers and phases of the fundamental mode and the third harmonic are shown in Figs. 5(a) and 5(b), respectively. Both distributions are symmetric with respect to the zero phase. For each mode in the state with a negative phase and in the state with a positive phase the values of photon numbers are the same. The maximum value of the photon number in the fundamental mode is realized together with the most probable value of the phase for this mode. The zero value of the phase is realized with the minimum value of the photon number. For the third harmonic the maximum and minimum values of photon number can be realized when this mode accepts the zero value of the phase.

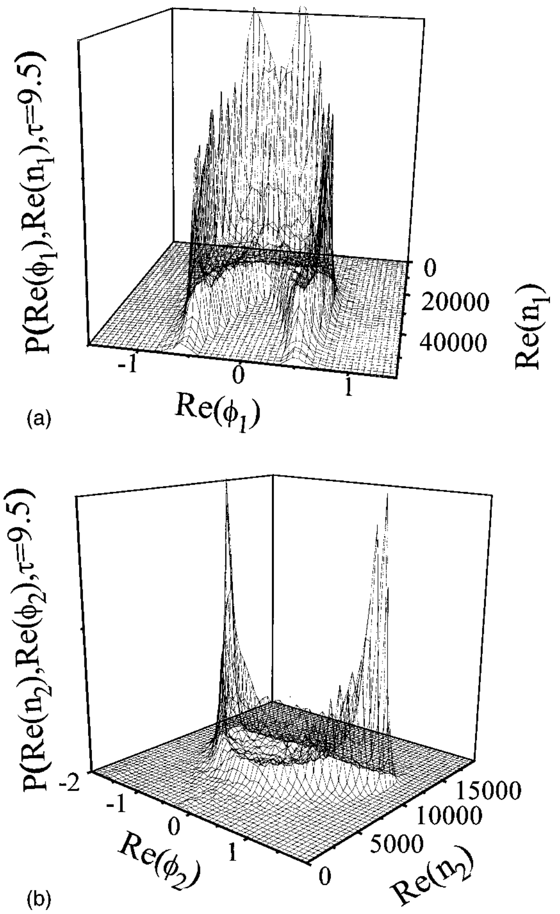

FIG. 5. Density of joint distribution of photon number and phase for the fundamental mode (a) and third harmonic (b) at the moment of time $\tau = 9.5$ and for values of parameters of Fig. 1. 100 000 trajectories.

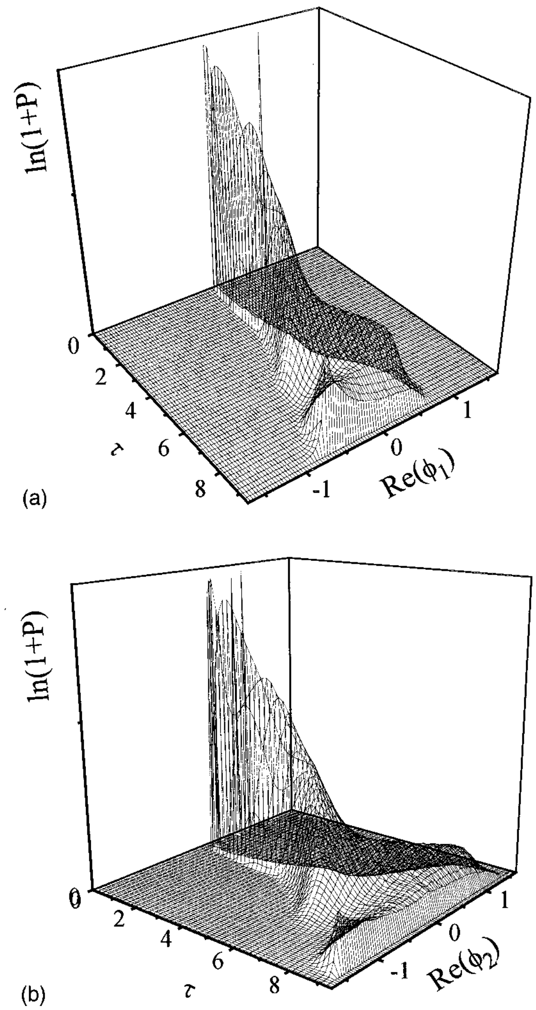

FIG. 6. Dynamics of distribution function of a phase for the fundamental mode (a) and third harmonic (b) in the case of coherent initial state of both modes. 100 000 trajectories.

IV. NONSTATIONARY SUPERPOSITION STATES

We now proceed with the calculation of the dynamics of the fundamental mode phase distribution function in the case of coherent initial states in both modes:

$$\alpha_j(0) = 1 + i, \quad \beta_j(0) = 1 - i \quad (i,j = 1,2). \tag{10}$$

Dynamics of this function is shown in Fig. 6(a). In this case the distribution function in the domain of large times has no stationary solution. The distribution function turns into the auto-oscillation regime. A similar behavior exhibits also the distribution function of the third-harmonic phase [see Fig. 6(b)]. For a brighter illustration of behavior of an optical system in the case of a coherent initial state the dynamics of the average phase of third harmonic is shown in Fig. 7. At the moments of time corresponding to the points *C*, the system is in superposition states. The density of the joint distribution of photon numbers and phases in these points coincides with the function presented in Fig. 5(b). At the moments of time corresponding to the points *N*, the system moves towards the single peak state with the negative most probable value of the phase. The density of the joint distribution of photon numbers and phases at these moments of time is shown in Fig. 8. At the moments of time corresponding to the points *P*, the system is in the state with a positive value of the phase. Thus the superposition states are destroyed with time and formed again in the system. A similar temporal behavior also exhibits the fundamental mode.

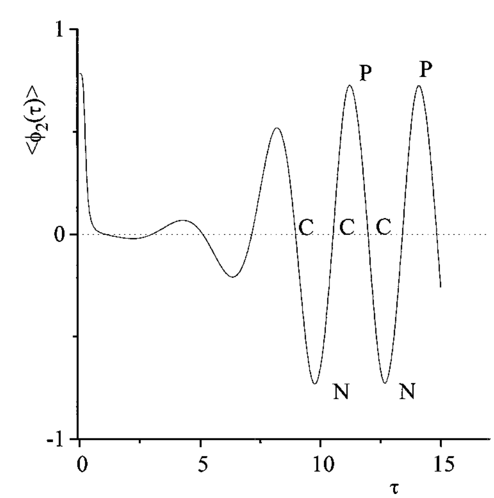

FIG. 7. Dynamics of the average phase for the third harmonic. 50 000 trajectories.

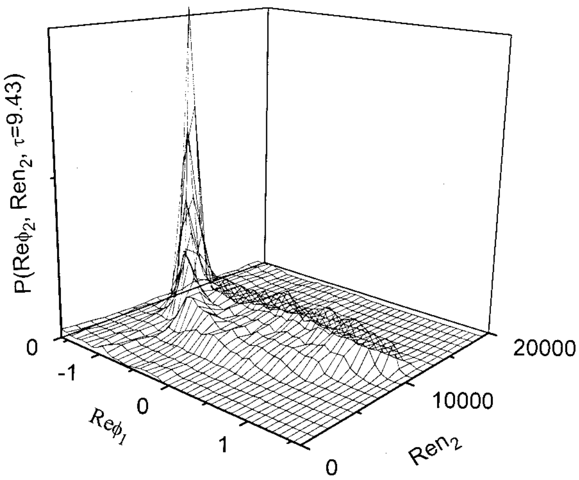

FIG. 8. Density of joint distribution of photon number and phase for the third harmonic at the moment $\tau=9.43$ and in the case of coherent initial state of both modes. 100 000 trajectories.

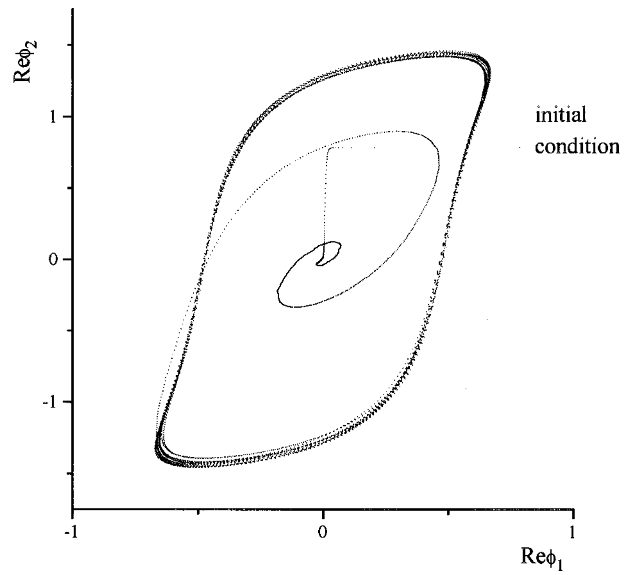

FIG. 10. A trajectory of optical system in the phase space (Re ϕ_1,Re ϕ_2). The trajectory is shown in the time interval $\tau=0-55$. Density of points is proportional to the stay time of the optical system in this area of phase space. A temporal interval between neighboring points is $\Delta\tau=0.01$.

V. PHASE MATCHING IN THIRD-HARMONIC GENERATION

In this section the function of the joint distribution of phases of the fundamental mode and third harmonic is investigated. This function at the moment $\tau=9.5$ and in the case of a Gaussian initial state in both modes (7) is shown in Fig. 9. The distribution density is calculated for the values of parameters as in Fig. 1. The function has two peaks which determine two states of the optical system. In either of these states the third harmonic and the fundamental mode accept identical signs of phases concerned with the phase of perturbation fields.

Such a state of an optical system has no classical analog, so this system is a quantum object. This state of the system can be termed a two-mode superposition state. The events in some domain surrounding the point (Re $\phi_1=0$, Re $\phi_2=0$) (this point determines the phase matching of modes in classical solutions) have zero probability of realization. As the quantum system approaches the bifurcation point from the right ($\varepsilon\rightarrow\varepsilon_{cr}$) the above-stated domain decreases and in the critical point we obtain a sharp peak at the value (Re $\phi_1=0$, Re $\phi_2=0$) instead of a dip around this value. The system becomes a classical object.

In Fig. 10 one trajectory of an optical system is shown in the phase space (Re ϕ_1,Re ϕ_2). In a certain very short time $\tau\ll 1$ after the interaction begins a classical matching of phases of interacting modes occurs in the cavity volume. A dense congestion of points around the values (Re $\phi_1=0$, Re $\phi_2=0$) corresponds to this matching. The density of points is proportional to a time of stay of the optical system in this domain of the phase space. After this the system transits to a state which has no classical analog. Two congestions of phase points determine the two states of the optical system.

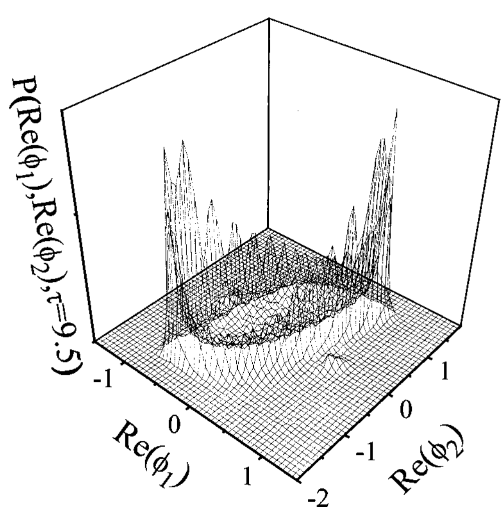

FIG. 9. Density of joint distribution of phases for the fundamental mode and third harmonic at the moment $\tau=9.5$ and in the case of initial Gaussian state of both modes. The function is calculated for values of parameters of Fig. 1. 100 000 trajectories.

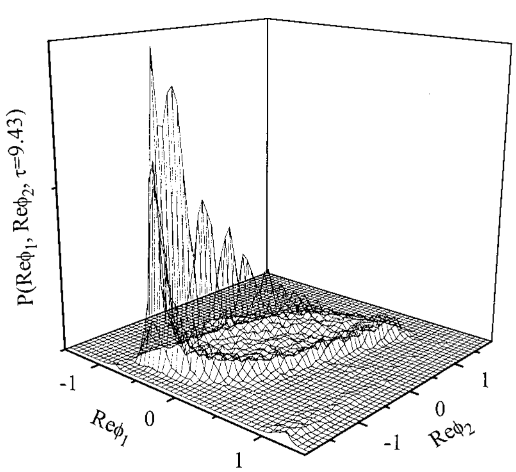

FIG. 11. Density of joint distribution of phases for the fundamental mode and third harmonic at the moment $\tau=9.43$ and in the case of coherent initial state of both modes. 100 000 trajectories.

In the case of a coherent initial state of both modes, Eq. (10), the distribution function turns at large times into the auto-oscillation regime. At the moments of time corresponding to the points C in Fig. 7 the distribution density coincides with the function of Fig. 9. For moments of time which correspond to the points N, this function is shown in Fig. 11. Here the system is located in one peak state where both phases accept negative values. At moments of time which correspond to the points P, the system is located in a state with positive values of phases of both modes.

Equations (2) for large times $\tau \gg 1$ can be solved numerically in instability region only for small nonlinearities $k^2 \ll 1$. For the large time intervals $\tau > 10$ jumps are appearing on the photon number trajectory leading to nonphysical peaks arising for the mean values of photon numbers.

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