cta physica slovaca vol. 46 No. 3, 221 – 230 June 1996

NONCLASSICAL EFFECTS IN $\chi^{(2)}$ MEDIA. CLASSICAL SIMULATIONS OF QUANTUM DYNAMICS[1]

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Received 31 May 1996, accepted 7 June 1996

We study the interaction between three light modes in $\chi^{(2)}$ media. In the degenerate as well as the nondegenerate case we can introduce two different types of phase space motion which we call phase stable and phase moving. We investigate the correspondence between the characteristics of the phase space motions in the classical and quantum domain. Using classical trajectories we construct the approximate quasiprobability distribution function in phase space which enables us to describe also quantum effects. This approach can be used to study the interaction of light for high photon numbers. The limitations of this approach are pointed out as well as the possibility to describe other nonlinear interactions of the light modes.

1. Introduction

The dynamics of light modes in nonlinear dielectrics was extensively studied since the sixties. The construction of the laser enabled theoretical as well experimental investigation of the behaviour of matter under extreme electromagnetic field power densities [1,2]. The basic theory came out soon after the publication of the first successful experiments and started so the field of classical and quantum nonlinear optics. The current spectrum of the processes in nonlinear optics includes processes like second and third

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0323-0465/96

harmonic generation, parametric down conversion, four-wave mixing to name a few. In particular the three wave interaction attracted great interest. It describes a process where from the pump mode we generate two other field modes namely the signal and the idler. Naturally also the reverse regime is possible where we generate through sum-frequency generation the pump mode. In practical implementations the modes are considered empty or highly excited (so called parametric processes). This simplifies considerably the description of the process, however it also inevitable leads to the neglect of certain effects.

In the following we look at certain effects in the nondegenerate as well as the degenerate three-wave interaction. The main attention is focused on the dynamics in phase space both on classical and quantum level. We show that using classical trajectories the approximate Wigner function in phase space can be constructed which describes also quantum effects.

2. Classical versus quantum description

The nondegenerate three-wave interaction is in the classical domain described by a set of equations for the field amplitudes and phases. We neglect any phase mismatch and assume exact resonance. The equations take the form [2,3,4] for the amplitudes

$$\begin{aligned} \frac{d}{d\zeta}u_s &= -u_i u_p \sin\theta; \\ \frac{d}{d\zeta}u_i &= -u_s u_p \sin\theta; \\ \frac{d}{d\zeta}u_p &= u_s u_i \sin\theta. \end{aligned} \tag{1}$$

and for the phases

$$\begin{aligned} \frac{d}{d\zeta}\varphi_s &= \frac{u_i u_p}{u_s}\cos\theta = \frac{\Gamma}{u_s^2}; \\ \frac{d}{d\zeta}\varphi_i &= \frac{u_s u_p}{u_i}\cos\theta = \frac{\Gamma}{u_i^2}; \\ \frac{d}{d\zeta}\varphi_p &= \frac{u_s u_i}{u_p}\cos\theta = \frac{\Gamma}{u_p^2}. \end{aligned} \tag{2}$$

In the equations we used the following variables: ζ is the scaled time $\zeta = \kappa t$ (κ is the coupling constant), θ is the phase difference defined as

$$\theta = \varphi_p - \varphi_i - \varphi_s. \tag{3}$$

The indices s, i, p refer to the particular wave, i.e., to signal, idler and pump,respectively. To solve this set of six equations one has to realize that the parameter Γ is an integral of motion [2]

$$\Gamma = u_s(\zeta)u_i(\zeta)u_p(\zeta)\cos\theta(\zeta). \tag{4}$$

what follows from the known equation of motion for the phase difference

$$\frac{d}{d\zeta}\theta = \frac{\cos\theta}{\sin\theta}\frac{d}{d\zeta}\ln(u_s\, u_i\, u_p). \tag{5}$$

In addition using the known constants of motion (only two are independent) [2,3]

$$m_1 = u_i^2 + u_p^2 \ , \ m_2 = u_s^2 + u_p^2 \ , \ m_3 = u_s^2 - u_i^2. \tag{6}$$

we write the solution for the squared field amplitude of the pump by means of Jacobian elliptic function as [the other amplitude can be obtained from the conserved quantities (6)]

$$u_p^3(\zeta) = u_{3a}^2 + (u_{3b}^2 - u_{3a}^2)sn^2[\sqrt{u_{3c}^2 - u_{3a}^2}(\zeta + \zeta_0), m], \tag{7}$$

where u_{3x}^2 are the roots of the cubic equation

$$x^3 - (m_2 + m_1)x^2 + m_1 m_2 x - \Gamma^2 = 0,$$

and the constant m equals

$$m = \frac{u_{3b}^2 - u_{3a}^2}{u_{3c}^2 - u_{3a}^2}.$$

The actual value of Γ does not only help to find the solutions for the amplitudes but can also serve for the natural classification of the phase motion. Knowing the solution for the amplitude we write the corresponding solution for the phase as

$$\varphi_x(\zeta) = \varphi_x(0) + \int_0^\zeta \frac{\Gamma}{u_x^2(\xi)}\,d\xi \tag{8}$$

In the case $\Gamma = 0$ the initial value of the phase $\varphi(0)$ does not change during the time evolution except the moments when the corresponding amplitude becomes zero $u_x = 0$. In this moment the phase can change by a factor of π. The phase space motion for this regime is realized on straight lines crossing the origin, i.e., the modes move radially. The realization of this regime can be achieved either by setting one of the initial field amplitudes to zero or adjusting the phase difference $\theta = \pm\pi/2$. This *phase stable* regime leads to the best possible energy conversion for given input intensities [as follows from the equations (1)].

The other regime - *phase moving* - corresponds to $\Gamma \neq 0$. Because the constant Γ is at any time nonzero each of the amplitudes will be nonzero and hence the individual phases of the modes can be obtained by a simple integration of the Eq.(8). Especially simple motion can be achieved in the *no-energy exchange* regime. Let us look for this once again closer at the Eqs. (1),(2) and (4). To fulfill the condition of no energy transfer we have to set the phase difference initially to $\sin\theta = 0$ and to choose the amplitudes according to the following condition

$$\frac{1}{u_s^2} + \frac{1}{u_i^2} = \frac{1}{u_p^2}. \tag{9}$$

When this relation holds for the initial amplitudes of the modes the particular phases evolve as

$$\varphi_x(\zeta) = \varphi_x(0) + \frac{\Gamma\zeta}{u_x^2}, \tag{10}$$

and the fastest phase motion is observable with the least excited mode. The points describing the mode evolve along closed circles, i.e. they, just rotate in phase space with constant angular velocity.

In the quantum domain the nondegenerate three wave mixing is described by the interaction Hamiltonian [4,5]

$$\hat{H}_{int} = \kappa(\hat{a}\hat{b}\hat{c}^\dagger + \hat{a}^\dagger\hat{b}^\dagger\hat{c}). \tag{11}$$

The three operators $\hat{a}$, $\hat{b}$ and $\hat{c}$ correspond to the annihilation operator of the signal, idler and pump mode.

The role of the preserved quantity Γ from the classical analysis is now taken over by the mean value of the interaction Hamiltonian which is integral of motion. This can be easily illustrated for instance using as initial inputs coherent states

$$|\psi(t=0)\rangle = |\alpha\rangle_s|\beta\rangle_i|\gamma\rangle_p, \tag{12}$$

with

$$\alpha = |\alpha|\exp(i\varphi_\alpha) \ , \ \beta = |\beta|\exp(i\varphi_\beta) \ , \ \gamma = |\gamma|\exp(i\varphi_\gamma). \tag{13}$$

With these input states the mean value of the interaction Hamiltonian takes the from

$$\langle\hat{H}_{int}\rangle = 2\kappa|\alpha||\beta||\gamma|\cos(\varphi_\alpha + \varphi_\beta - \varphi_\gamma). \tag{14}$$

In strict analogy to the classical case we can distinguish two types of phase space motion. For $\langle\hat{H}_{int}\rangle = 0$ we have the ***phase stable*** motion, in the case $\langle\hat{H}_{int}\rangle \neq 0$ we have the ***phase changing*** motion.

The phase stable motion cover very important quantum-mechanical regimes like sum-frequency generation or parametric down-conversion leading to the generation of a two-mode squeezed vacuum. For visualization of the phase-space motion the quasiprobability distributions such as the Husimi Q- and Wigner W-function[5] can be used [6]. In the phase stable regime the initial (coherent state) distribution moves radially - with its center along straight line through the center. As time elapses the initial distribution is distorted due to the fine details of the quantum dynamics.

The phase-changing regime corresponding to $\langle\hat{H}_{int}\rangle$ requires all modes to be initially excited. In the phase space the centers of blobs (e.g. corresponding to initially coherent states) move along quite complicated lines corresponding to the classical solutions. In the discussion about classical solutions we pointed out a special classical regime with no energy exchange between the modes [see Eq.(9)]. In the quantum regime this condition cannot guarantee the complete suppression of the energy flow between the modes. However, it makes the effect small. In the initial moments the energy exchange is an effect of third order in time and just on the long time scale a visible energy transfer between the modes can be seen. In this particular case the phase space motion proceeds along circles with the typical Kerr-like deformation of shape due to quantum phase spreading.

[5] for their definitions see, e.g., the paper by J.Janszky et al. in this issue of *Acta Physica Slovaca*

Let us notice that this "no-energy exchange" regime is interesting owing to the possibility to produce strongly sub-Poissonian light. The degree of sub-Poissonian character is defined using the Mandel's q-parameter [7]

$$q_x = \frac{\langle(\hat{x}^\dagger\hat{x})^2\rangle - \langle\hat{x}^\dagger\hat{x}\rangle^2}{\langle\hat{x}^\dagger\hat{x}\rangle} - 1.$$

With a proper phase and intensity adjustment we can suppress dynamically the energy transfer between the modes, however we still can manipulate the fluctuations of the photon number either by a proper choice of initial states or their entanglement [4,8,9]. With an initial Kerr-state ansatz [10] for the signal we can obtain significant sub-Poissonian light [9]. The degree of sub-Poissonian character is limited in this case by the phase space motion. To enhance the effect further we would need to keep the initially adjusted phase difference which is unfortunately changed during the time evolution.

The classical version of the degenerate three wave interaction starts from the coupled equations for the fundamental v_s and the second harmonic v_p wave (assuming exact resonance and phase matching)

$$\begin{aligned} \frac{d}{d\zeta}v_s &= -2v_s v_p \sin\theta; \\ \frac{d}{d\zeta}v_p &= v_s^2\sin\theta; \end{aligned} \tag{15}$$

$$\begin{aligned} \frac{d}{d\zeta}\varphi_s &= 2v_p\cos\theta = \frac{2\Gamma_2}{v_s^2}; \\ \frac{d}{d\zeta}\varphi_p &= \frac{v_s^2}{v_p}\cos\theta = \frac{\Gamma_2}{v_p^2}. \end{aligned} \tag{16}$$

The phase difference $\theta = \varphi_p - 2\varphi_s$ satisfies the equation

$$\frac{d}{d\zeta}\theta = \left(\frac{v_s^2}{v_p} - 4v_p\right)\cos\theta = \left(\frac{1}{v_p^2} - \frac{4}{v_s^2}\right)\Gamma_2. \tag{17}$$

The solution of the written set of equations can be obtained along almost identical lines as in the nondegenerate case, i.e., by employing the existence of the integral of motion $\Gamma_2 = v_s^2 v_p\cos\theta$. As a consequence we can introduce the same classification for the phase-space motion. Let us turn to the quantum picture.

The quantum description of the second harmonic generation is given by the effective interaction Hamiltonian

$$\hat{H}_2 = \kappa_2(\hat{a}^2\hat{c} + \hat{a}^{2\dagger}\hat{c}) \tag{18}$$

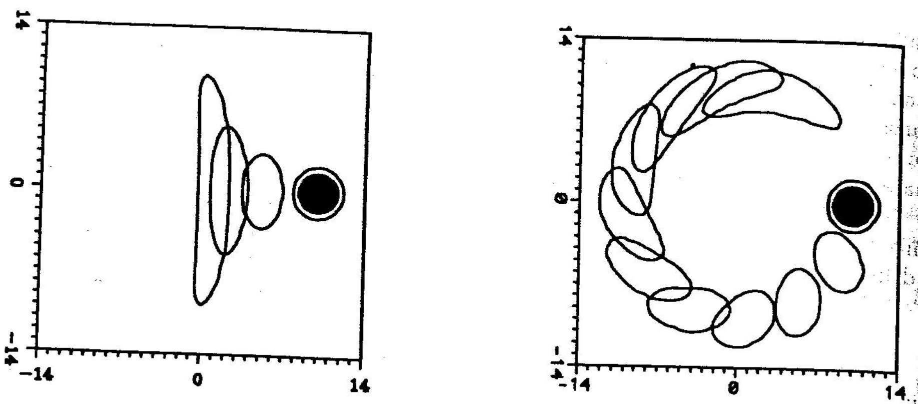

Fig. 1. The Husimi Q-function (contour) of the fundamental wave for the initial state (16) with $|\alpha| = 10$, $|\gamma| = 5$. (a) $\varphi_\gamma = -\pi/2$ corresponds to the phase stable motion - in radial direction; (b) $\varphi_\gamma = 0$ is similar to the classically no-energy exchange regime characterized by rotation in phase space.

The mean value of the interaction Hamiltonian $\langle \hat{H}_2 \rangle$ is again a conserved quantity which represents a quantum analogue of the classical integral of motion Γ_2. It enables us to distinguish two basic forms of phase-space motion and to connect the classical phase-motions and their quantum counterparts. The two most important phase-space motions are illustrated in Fig. 1 in terms of the Husimi Q-function of the fundamental mode (signal). Initially both modes are prepared in coherent states

$$|\alpha\rangle_s |\gamma\rangle_p, \tag{19}$$

with $\alpha = |\alpha| e^{i\varphi_\alpha}$ and $\gamma = |\gamma| e^{i\varphi_\gamma}$. Namely, the amplitudes are $|\alpha| = 10$, $|\gamma| = 5$ and the phases $\varphi_\alpha = 0$, $\varphi_\gamma = -\pi/2, 0$. Fig. 1a with $\varphi_\gamma = -\pi/2$ corresponds to the phase stable motion (i.e., the radial motion of the centre of the Q-function in phase space). It is seen that considerable amplitude squeezing can be achieved. The classical no-energy exchange regime corresponds to $\varphi_\gamma = 0$ what is reflected in quantum domain by rotation of the Q-function in phase space - see Fig. 1b. It is worth to notice that in the $\chi^{(2)}$ nonlinearity we can obtain in the "no-energy exchange regime" the behaviour of the modes typical for Kerr-like medium which is associated with $\chi^{(3)}$ nonlinearity [10].

3. Quantum description via classical trajectories

We showed, that the classification of the classical phase space motion can be reformulated in a close analogy also for the quantum picture of the three-wave dynamics. It is of interest to know, to what extend we can use the classical solutions of the three-wave interaction in the quantum domain. In a simpler formulation of the problem, namely in the so called parametric approximation, the solution is known. In such regimes one of the modes is highly excited and can be treated as a classical field (the corresponding

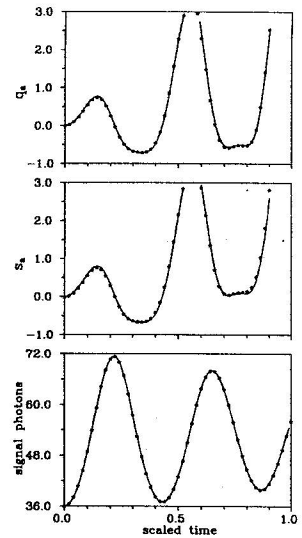

Fig. 2. The signal mean photon number, quadrature squeezing and Mandel's q parameter for the case of sum-frequency generation with $|\psi(0)\rangle = |\alpha = 9\rangle|\beta = 5\rangle|\gamma = 0\rangle$. The exact solution is plotted by solid line, Wigner function approach by $\star$ and Q function by Δ.

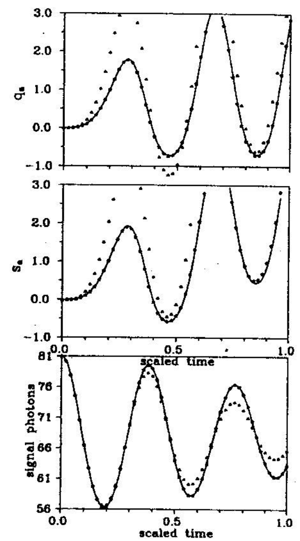

Fig. 3. The same as in Fig. 2 but for the difference frequency generation with $|\psi(0)\rangle = |\alpha = 6\rangle|\beta = 0\rangle|\gamma = 6\rangle$.

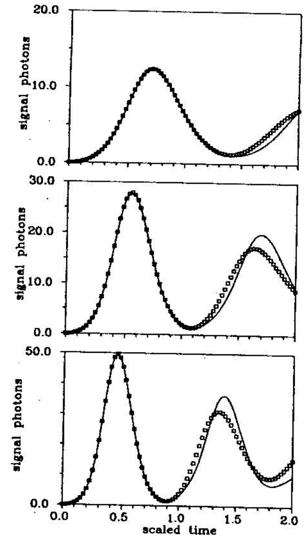

Fig. 4. The signal photon number for down-conversion with $|\psi(0)\rangle = |\alpha = 0\rangle|\beta = 0\rangle|\gamma = 4, 6, 8\rangle$. The exact solution is shown with a solid line and the approximate one with squares. The validity of the approximation is not enhanced with increased intensity.

operators are replaced by complex numbers). It means that the model Hamiltonian degenerates to a quadratic form. It was pointed out by Mollow and Glauber [6] that in the case of quadratic Hamiltonians we can use the classical solutions to obtain the exact quantum evolution. The trick is to use as arguments of the Wigner function the classical solutions. Namely, for the parametric three–wave mixing with strong pump, i.e., when c mode is treated classically in Eq.(11),

$$W[\alpha,\beta;t] = W[\alpha_0(\alpha,\beta,t),\beta_0(\alpha,\beta,t);0], \tag{20}$$

where $\{\alpha_0(\alpha,\beta,t),\beta_0(\alpha,\beta,t)\}$ is trajectory in classical phase space (for signal and idler) which at time t approaches point $\{\alpha,\beta\}$. In other words, the value of the Wigner function at time t and the point $\{\alpha,\beta\}$ in phase space is obtained evolving this point backwards in time according to classical equations of motion and taking the value of the Wigner at $t = 0$ for corresponding initial point $\{\alpha_0,\beta_0\}$.

To see more explicitly that Eq.(20) is valid let us consider the quantum Liouville equation for Wigner function for a one-mode system with canonically conjugate variables q (position) and p (momentum):

$$\frac{\partial W}{\partial t} = \underbrace{-\frac{\partial H}{\partial p}\frac{\partial W}{\partial q} + \frac{\partial H}{\partial q}\frac{\partial W}{\partial p}}_{\text{"classical terms"}} \underbrace{- \frac{\hbar^2}{24}\frac{\partial^3 H}{\partial q^3}\frac{\partial^3 W}{\partial p^3}}_{\text{"quantum terms"}} + O(\hbar^4). \tag{21}$$

It is evident that the quantum Liouville equation is fully equivalent to the classical Liouville equation only for *quadratic* Hamiltonians. In such situation it is possible to describe by classical trajectories even initially negative Wigner functions [see Eq.(20)].

We applied the Glauber's philosophy to the case of all three modes excited (nondegenerate three-wave interaction) [11] taking the approximate Wigner function

$$W[\alpha,\beta,\gamma;t] = W[\alpha_0(\alpha,\beta,\gamma,t),\beta_0(\alpha,\beta,\gamma,t),\gamma_0(\alpha,\beta,\gamma,t);0] \tag{22}$$

and neglecting thus "quantum terms" in the corresponding quantum Liouville equation [see (21)]. In practice, for three modes we should browse through six-dimensional phase space what is not practically possible. Therefore Monte Carlo methods have to be adopted with the *importance sampling*. In other words, the quantum dynamics in phase space is simulated within the *classical* phase space using an initial ensemble of phase–space points each representing a classical initial configuration and evolving along a classical trajectory. The initial probability distribution in the classical phase space reflects directly the quantum fluctuations being chosen equal to an initial quasiprobability distribution like Husimi (Q) or Wigner (W) function.

Some numerical results are presented in Figs. 2-4. The three figures cover the cases of sum-frequency generation ($|\psi(0)\rangle = |\alpha\rangle|\beta\rangle|0\rangle$ – Fig.2), difference frequency generation ($|\psi(0)\rangle = |\alpha\rangle|0\rangle|\gamma\rangle$ – Fig.3) and down-conversion ($|\psi(0)\rangle = |0\rangle|0\rangle|\gamma\rangle$ – Fig.4). The figures clearly demonstrate that there are certain limits for the applicability of the given method. Even though there is still an excellent agreement between the mean photon numbers (the results by the Wigner function are shown with stars), the higher moments represented by the Mandel's q-parameter and quadrature squeezing (for definition see [4,9]) show already some deviation when compared with the exact quantum-mechanical calculation (shown as solid lines in Figs. 2-4). However, it is remarkable that the validity of such an approach goes beyond the short time approximation, i.e., it covers at least one quasiperiod of the energy flow between the modes. The other limitation is the dependence on the initial state. In Fig. 4 the signal photon number is shown for various values of γ. Here the approximation holds only till the second reversal of the energy flow. In Fig. 2 we included also the calculation using the classical simulation of the Q function and afterwards we calculated the shown parameters. Even though qualitatively the Q function shows a good agreement what the phase-space dynamics concerns [12,13], it does not stand the quantitative test beyond the initial moments of time. In other words, the Wigner method seems to be about as nearly classical as it is possible for a full quantum theory.

Conclusions

We showed, that the dynamics of three waves in $\chi^{(2)}$ media can be in a natural way classified in the classical as well as quantum domain using a proper integral of motion for the degenerate as well as nondegenerate two-photon down-conversion. In the classical case the classification is done using the constant Γ and in the quantum case by the mean value of the interaction Hamiltonian $\langle \hat{H}_{int}\rangle$. In the case when these constants equal to zero we deal with the phase stable regime. Apart from a possible phase jump by π the phases of the modes stay on their initial values. This regime is also associated with the best energy transfer between the modes. In the case of nonzero constants Γ and $\langle \hat{H}_{int} \rangle$ we deal with the phase changing regime. In this case the individual phases as well as the phase difference change. Let us stress that for a special initial choice of amplitudes and phases we can establish a regime with no energy exchange in the classical domain and with a feeble exchange in the quantum domain. In the quantum formulation of the considered process with $\chi^{(2)}$ nonlinearity such an initial state mimics the evolution typical for a Kerr-like medium.

The classical dynamics can be effectively used not only for the classification of phase-space motion but also for classical simulations of the quantum dynamics. The knowledge of the phase-space trajectories can be used in evolving the initial quantum Wigner distribution along classical paths. In this way we obtain not only a good qualitative insight into the quantum dynamics on a relatively long time scale but also a tool to calculate quantum averages especially for large initial intensities. It is particularly interesting that this approach is not restricted to coherent inputs but it can be applied also for other initial states even for those which are characterized by negative Wigner functions. In the parametric regimes (quadratic Hamiltonians) such an approach is exact, in the case of three waves the method can fail quantitatively after the first reverse of the energy exchange, i.e., in a time region beyond the scope of any perturbative approach. The classical trajectories can be used for description of the quantum dynamics also for other nonlinear processes, e.g., four wave mixings.

Acknowledgements The support to one of us (G.D.) granted by the Max-Planck Society is gratefully acknowledged.

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