Classical-to-quantum transition in multimode nonlinear systems with strong photon-photon coupling

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With advanced micro- and nano-photonic structures, the vacuum photon-photon coupling rate is anticipated to approach the intrinsic loss rate and lead to unconventional quantum effects. Here, we investigate the classical-to-quantum transition of such photonic nonlinear systems using the quantum cluster-expansion method, which addresses the computational challenge in tracking large photon number states of the fundamental and harmonic optical fields involved in the second harmonic generation process. Compared to the mean-field approximation used in weak coupling limit, the quantum cluster-expansion method solves multimode dynamics efficiently and reveals the quantum behaviors of optical parametric oscillations around the threshold. This work presents a universal tool to study quantum dynamics of multimode systems and explore the nonlinear photonic devices for continuous-variable quantum information processing.

I. INTRODUCTION

Nonlinear optics has been exploited in abundant classical and quantum optics applications since the advent of lasers [1, 2]. Under the current theoretical framework, the simplest approach to describe a coherent optical field is by characterizing it with only one parameter, i.e., the field amplitude, with the system dynamics governed by a set of nonlinearly coupled equations among modes of different amplitudes. This treatment, known as the mean-field approximation (MFA) [3], however neglects the influence of the quantum fluctuations. In a more rigorous framework, the optical fields are treated as Gaussian states for which the mean-field amplitude and second-order correlations are assumed to be complete to describe the system [4, 5]. Then, the quantum fluctuations and correlations of optical fields can be derived by calculating the covariance matrix. In the past decades, the MFA and Gaussian-state approximation have been widely applied in quantum optics and successfully predict a variant of phenomena, including the squeezing [6], continuous-variable entanglement [7], and the thermal dynamics of mechanical resonators [8, 9].

In conventional nonlinear optics systems, the nonlinear coupling strengths between optical modes are much weaker than the dissipation rates, therefore the higher-order correlations between modes are negligible due to the strong decoherence, and the Gaussian state approximation is accurate. As the fabrication technique and material improves, the photon-photon coupling in the nonlinear system can be greatly enhanced, to the point that the single-photon nonlinearity becomes appreciable in photonic integrated circuits. The coupling strength to dissipation rate ratio \( g/\kappa \) has been greatly boosted during the last decades [10], using a microresonator made by gallium arsenide (GaAs) [11], aluminum nitride (AlN) [12], indium gallium phosphone (InGaP) [13], lithium niobate (LN) [14–16] and etc. For example, Lu et al. demonstrated a \( g/\kappa \) ratio over 1 % in a periodically poled lithium niobate microring resonator [17], suggesting significant nonlinear effects at the level of tens photons. At this high nonlinearity limit, the Gaussian state approximation no longer holds. With even larger \( g/\kappa \), the nonlinear system is predicted to exhibit atom-like features [18, 19], thus significant quantum effects arise under excitation at the single-photon level [20, 21]. It is intriguing to explore the classical-to-quantum transition in this new regime where \( g/\kappa \) approaches unity and the conventional treatment of high amplitude bosonic modes under MFA is no longer valid, and the quantum master equation with truncated Fock-state dimension becomes inefficient.

In this Letter, the classical-to-quantum transition of nonlinear \( \chi^{(2)} \) processes is investigated based on the quantum cluster-expansion (QCE) approach. In particular, we focus on degenerate \( \chi^{(2)} \) interactions, including second-harmonic generation and optical parametric oscillation at different nonlinear coupling rates and pump powers. The numerical results show the deviation of the mean photon numbers and the quantum statistics from the predictions by the classical theory, and manifest the classical-to-quantum transition when increasing the \( g/\kappa \) and the pump power. We developed the code for generating recursive QCE to arbitrary orders with an arbitrary number of modes. By comparing with the conventional numerical approaches based on master equations of truncated Hilbert space, the validity of QCE is verified and shows a \( 10^4 \) times speedup under excitation of only 400 photons. Our approach is efficient for solving the problems with large intracavity photon numbers and also moderate \( g/\kappa \) ratio, and could be extended to study the quantum behaviors of other complex nonlinear optics systems.

II. PRINCIPLE OF QCE

In resonance-enhanced nonlinear photonics processes such as three-wave mixing and frequency comb generation in microresonators [22], there are multiple optical resonances simultaneously satisfying the energy and phase matching conditions. These modes are generally described by the bosonic annihilation...
of Otonian that consists of only 2nd-order clusters, the evaluations should depend on the higher order clusters \[\text{Fig. 1(c)}\], and 3rd or higher order clusters, the dynamics of 2nd-order clusters \[\text{covariance matrix}\]. However, for the system that involves quantum state evolution can be completely described by the first- and second-order clusters \[\text{Fig. 1(b)}\], and the system initial states are Gaussian states, the expectation values of the higher-order clusters can be expressed in terms of system initial states are Gaussian states, the expectation values of the higher-order clusters can be expressed in terms of the Hamiltonian of the system \[3\]. If all the environment modes and the Hamiltonian \(H\) and higher-order nonlinear interactions, i.e. the interaction operators \(O_j\), where \(j\) labels individual mode, and any given system operator could be written as the product of a cluster of mode operators \(A = \prod_{j,k} O_j^{(m_j)} O_k^{(n_k)}\), which is a \(M\)-th order operator with \(M = \sum m_j + \sum n_k\). For an open quantum system, the dynamics of an operator \(A\) follows the master equation \[1\]

\[
\frac{d}{dt} \rho = -i[H, \rho] + \sum_j \kappa_j \mathcal{L}_{d_j} [\rho],
\]

where \(\rho\) is the density matrix and \(H\) is the Hamiltonian of the system, \(\kappa_j\) is the dissipation rate and \(\mathcal{L}_{d_j} [\rho] = 2d_j \rho d_j^\dagger - d_j^\dagger d_j \rho - \rho d_j^\dagger d_j\) is the Lindblad operator for a jump operator \(d_j\). For example, in the case of a reservoir with near-zero thermal excitation, the amplitude dissipation of individual modes is \(\kappa_j \mathcal{L}_{d_j} [\rho]\). The expectation value of an operator \(\langle A \rangle = \text{Tr} \{ A \rho \}\) also follows the master equation \[2\]

\[
\frac{d}{dt} \langle A \rangle = i \frac{\hbar}{\kappa} [H, A] + \sum_j \kappa_j \langle \mathcal{L}_{d_j}' [A] \rangle.
\]

The master equation shows that the dynamics of the expectation value of the \(M\)-th order cluster \(A\) is directly coupled to operators \([H, A]\) and also \(\mathcal{L}_{d_j}' [A]\). For a simple bilinear Hamiltonian that consists of only 2nd-order clusters, the evaluations of \(O_j\) only depends on the first order clusters, thus the master equations of the system become an array of closed-form linear equations of \(O_j\) and \(O_j^\dagger\), as shown by Fig. 1(a). The expectations of all 2nd-order clusters then can be derived based on this set of linear equations, which further gives the covariance matrix of the system \[3\]. If all the environment modes and the system initial states are Gaussian states, the expectation values of the higher-order clusters can be expressed in terms of the first- and second-order clusters \[\text{Fig. 1(b)}\], and the system quantum state evolution can be completely described by the covariance matrix. However, for the system that involves \(\chi^{(2)}\) and higher-order nonlinear interactions, i.e. the \(H\) consists of 3-rd or higher order clusters, the dynamics of 2nd-order clusters should depend on the higher order clusters \[\text{Fig. 1(c)}\], and the evolution of all clusters in general cannot be obtained in a closed form. For example, for \(H = \hbar a^\dagger a (b + b^\dagger)\), the evolution of the expectation value of \(a\) is governed by

\[
\frac{d}{dt} \langle a \rangle = -i(\langle a(b + b^\dagger) \rangle - \kappa_a \langle a \rangle),
\]

which requires the values of \(\langle ab \rangle\) and \(\langle ab^\dagger \rangle\). Repeating the same process for \([H, A], A = \{ab, ab^\dagger\}\) leads to an infinite hierarchy that \(\langle A \rangle\) is expressed by an infinite set of higher-order operators.

Two approaches can be applied to address this divergence of high-order operators: (i) MFA, which neglects the fluctuations of strong fields and replaces them by complex numbers, thus reducing the order of clusters. In the classical limit, all operators are replaced by complex numbers, resulting in MFA shown in Fig. 1(a). (ii) Fock space truncation (FST). When the number of excitations in bosonic modes are restricted, the quantum state could be represented in the Fock basis with a finite dimension, and then the master equation could be solved numerically. Although both approaches are widely adopted in quantum optics studies, they are not applicable to a system with moderate nonlinearities and strong drives.

Therefore, we revert to solve the original master equation with the QCE approach. In practice, it is unrealistic to track the expectation values of an infinite set of operators to evaluate the expectation value \(\langle A \rangle\). Although the number of clusters involved for a complete system dynamics generally diverges, the QCE can be solved approximately or even analytically by truncating the order of QCE, i.e. setting the high order clusters as zero. Such treatment has been established in quantum chemistry \[23\] and semiconductor systems \[24\] as well as Bose-Einstein condensates beyond the mean-field theory \[25\]. Different from these previous studies, the order of the hierarchy of multimode nonlinear photon system depends on the specific nonlinear processes involved. At large mode number and orders, the expansion of the clusters increases drastically, and it is intractable to directly write down all the equations for clusters and solve them analytically.

The high-order correlation is directly related to the nonlinear coupling rate \(g\). In the weak-coupling limit \(g \ll \kappa\), the correlation between different operators can be neglected. The expectation values of \(N\)-th order operators can be directly factorized to the product of 1st order operators by

\[
\langle \hat{N} \rangle \approx \prod_{j=1}^{N} \langle O_j \rangle,
\]

which is the main assumption of the MFA. At this limit, the photonic modes are treated as harmonic oscillators and the optical fields can be approximated to coherent states, as has been adopted by most experiments. The system dynamics is described by a set of nonlinearly coupled equations only containing the expectation values of 1st order operators \[\text{Fig. 1(a)}\], while the tiny quantum fluctuations around the mean-field \(\langle O_j \rangle\) is neglected. As the \(g/\kappa\) ratio increases, the strong anharmonicity leads to the distortion of the quantum state from the coherent state, in which case the quantum correlation becomes
significant so that the high-order correlation can not be directly factorized to the product of single-order operators represented by Eq. (4). For example, a 2nd order operator can be written as \( \langle 2 \rangle = \langle 1 \rangle \langle 1 \rangle + \Delta \langle 2 \rangle = \langle 2 \rangle_s + \Delta \langle 2 \rangle \), where \( \langle 2 \rangle_s \) represents the MFA approximation by Eq. (4) and \( \Delta \langle 2 \rangle \) indicates the purely correlated part. Likewise, the factorization of a \( N \)th order cluster reads

\[
\langle \hat{N} \rangle = \langle \hat{N} \rangle_s + \langle \hat{N} - 2 \rangle \Delta \langle 2 \rangle + \langle \hat{N} - 4 \rangle \Delta \langle 2 \rangle \Delta \langle 2 \rangle + \ldots \Delta \langle \hat{N} \rangle = (O_a) \langle \hat{N} - 1 \rangle + \Delta (O_a \hat{1}) \langle \hat{N} - 2 \rangle + \Delta (O_a \hat{2}) \langle \hat{N} - 3 \rangle + \ldots \Delta \langle \hat{N} \rangle,
\]

where each product term presents one factorization and is summed over all indistinguishable combinations. \( \hat{1} \) denotes all the possible \( i \)th order cluster within the \( N \)th order cluster. And \( \hat{N} - i \) represents the factorization of the remaining \( \langle N - i \rangle \)th order cluster.

Based on Eqs. (2) and (5), the dynamics of a nonlinear system can be implemented following the \( M \)th order QCE:

1. To get the expectation value of \( \langle \hat{N} \rangle \), submit \( \langle \hat{N} \rangle \) to Eq. (2) and one gets its relation with \( \langle H, \hat{N} \rangle \) and operator \( \langle \hat{1} \rangle \) of other orders.
2. Following the \( M \)th truncation that all \( \Delta \langle \hat{N} \rangle = 0 \) for \( N \geq M \), \( \langle \hat{N} \rangle \) and \( \langle \hat{1} \rangle \) can be factorized according to Eq. (5).
3. Repeat Steps 1 and 2 for any cluster that appears in Step 2 until no new clusters are generated. In this way, we arrive at a set of nonlinear coupled equations involving clusters up to \( M \)-th orders.

We implement an open-source package to automatically with tree based symbol system to complete the above procedure. The technical details about the code are offered online and the codes are available in Ref. [26].

III. CLASSICAL-TO-QUANTUM TRANSITION OF \( \chi^{(2)} \) INTERACTION

We apply the QCE approach to investigate the classical-to-quantum transition of a signature quantum nonlinear optical system — degenerate \( \chi^{(2)} \) interaction — involved in the second-harmonic generation (SHG) and optical parametric oscillation (OPO). For the phase-matched degenerate \( \chi^{(2)} \) interaction between modes \( a \) and \( b \), the Hamiltonian can be written as [22, 27]

\[
H = \omega_a \hat{a}^\dagger \hat{a} + \omega_b \hat{b}^\dagger \hat{b} + g \left( \hat{a}^{\dagger 2} \hat{b} + \hat{a} \hat{b}^{\dagger 2} \right),
\]

where \( \hat{a} \) (\( \hat{b} \)) and \( \hat{a}^\dagger \) (\( \hat{b}^\dagger \)) are the annihilation and creation operator of the fundamental (second-harmonic) mode with \( \omega_a(b) \) being the corresponding resonant frequency. \( g \) is the nonlinear coupling strength that depends on the material and cavity geometry.

Figure 2 summarizes the results for SHG, under a coherent drive on mode \( a \) as \( E \left( \hat{a}^\dagger e^{-i\omega_d t} + a e^{i\omega_d t} \right) \), with \( E \) is the drive amplitude and \( \omega_d = \omega_a \) is the frequency of the drive resonant with mode \( a \). To validate the QCE, we compare the system evolution modeled by three different approaches. The master equation provides the most rigorous solution as long as the FST has the truncated dimension large enough. Due to the limited computation resource, we take the truncation dimension as 40 and 20 for mode \( a \) and mode \( b \), respectively. For comparison, the QCE is solved with the 4th-order truncation of clusters. The classical model with MFA \( \langle \hat{a}^\dagger \hat{b} \rangle = \langle \hat{a}^\dagger \rangle \langle \hat{b} \rangle \) is also evaluated, and we note that the MFA is actually the 1st-order QCE. For \( E = 6 \) and \( g = 0.4 \), with initial vacuum state, the dynamics of photon numbers in the two modes show excellent agreement between QCE and FST. However, the results of MFA deviate from the other two approaches, indicating the non-negligible quantum correlations between modes exist for \( g/\kappa \sim 0.4 \) as such effects could not be captured by the MFA. Additionally, the steady-state populations in two modes are studies for various \( g \) and \( E \). Figures 2(c) and (d) show that the MFA and QCE start to deviate when \( g \) exceeding 0.1, indicating a cross-over from classical to quantum regime. Similar behavior is shown when increasing \( E \) for a fixed \( g = 0.2 \), as shown in Figs. 2(e) and (f). Furthermore, MFA predicts a classical threshold above which will lead to self-pulsing as

![FIG. 2. Quantum-to-classical transition behaviors of second-harmonic generation.](image-url)
for $E < E_c$. Even though semi-classical modification can be introduced to MFA to explain the spontaneous parametric oscillation, it is still difficult to predict the system behavior precisely when the drive amplitude is around the threshold. For both SHG and OPO, MFA is consistent with other approaches at the weak coupling limit, thus validating the coupled-mode theory widely adopted for nonlinear optical systems with $g/\kappa \ll 1$. For OPO far above threshold ($E$ and $g$ are large enough), the photon number in the fundamental mode is much larger than the second-harmonic mode, thus their quantum correlation can be safely neglected i.e. $\Delta(2) \approx 0$. In this case, QCE and MFA agree with each other.

Besides offering more accurate prediction of the photon numbers, another prominent advantage of the QCE is its ability to track the quantum statistics of the optical fields when the nonlinear optical system transits from weakly anharmonic to strongly anharmonic regime. In quantum optics, the second-order self-correlation function

$$g^{(2)} = \frac{\langle O^\dagger O^\dagger OO \rangle}{\langle O^\dagger O \rangle^2}$$

is often used to quantify the quantum statistics of a mode $O$. It requires the operator cluster to be truncated at least to the 2nd-order, whereas MFA could not track the correlation functions since $\langle O^\dagger O^\dagger OO \rangle = \langle O^\dagger O \rangle^2$ for 1st order truncation. In contrary to the coupled-mode equations by MFA, which can be transformed to $g/\kappa$-invariant form [29], the quantum correlation function is coupled with the mean-fields in QCE, and thus depends on the pump power and $g/\kappa$. It is worth noting that the QCE also enables the calculation of arbitrary high-order quantum correlation functions by choosing an appropriate truncation order.

Figure 4 shows $g^{(2)}$ as a function of the coupling strength $g$ and the driving strength $E$. For SHG, both the $g^{(2)}$ function of the fundamental and SH mode are smaller than 1 when $g$ is
large and $E$ is small, revealing the photon anti-bunching due to the significant quantum photon blockade effect [20, 21]. As the driving strength $E$ becomes large, the $g^{(2)}$ function tends above 1, which corresponds to the bunching effect and can be attributed to optical bistability and bifurcation [30, 31]. The OPO case [Fig. 4(c)-(d)] also shows pump power-dependent $g^{(2)}$ function, which indicates the classical-to-quantum transition of OPO under a strong pump as the value of the $g^{(2)}$ function diverges from 1.

**IV. PERFORMANCE ANALYSIS**

It is of great importance to gain further insights into the performance of QCE as the nonlinear optical system transitions from weakly harmonic ($g/\kappa \ll 1$) to strongly anharmonic ($g/\kappa \gg 1$) regime. Figure 5 shows the performances of QCE for SHG and OPO with different QCE truncation orders. The solid line is obtained by solving the master equation in the Fock state basis and is used as a reference. For both SHG and OPO, the deviations of the QCE results become large with the increase of the coupling strength $g$, indicating an increased high-order quantum correlation. It is anticipated that it is more accurate to treat the nonlinear system as multilevel atoms when $g/\kappa \gg 1$ [19]. By expanding the operator clusters to higher-order, the results of QCE, as shown by the dashed curves with orders of 2, 4, and 6, converge to the result of the FST. However, as the order of expansion increases, a much larger number of clusters are involved in the nonlinear coupled equation, and lead to exceptionally high computational complexity, especially for $g \gg \kappa$.

Therefore, the potential advantage of the QCE approach is discussed by comparing its time consumption and computational complexity with other approaches. In a nutshell, Fig. 6(a) offers a qualitative illustration of the applicable parameter region for the three methods discussed in this paper. The MFA is efficient and accurate for very weak nonlinearity with $g/\kappa \ll 1$. For the FST, the density matrix contains the full information of the optical fields with a finite Fock space dimension, and can be used for the calculation of the expectation value of operators with arbitrarily high order. Thus, the FST method is particularly powerful for $g/\kappa \gg 1$, since only a small amount of photon number states is enough to capture the system’s behavior due to the strong anharmonicity, but is limited to very few modes. The QCE is more suitable for nonlinear systems with moderate nonlinearity $g/\kappa \lesssim 1$, and its superiority is particularly significant for strong pump power and large mode number. For example, in Fig. 2 with $g/\kappa = 0.2$, a 4-th order QCE is enough to predict the photon numbers of both modes with high precision. The star marked near the curve of QCE (Fig. 2(d)) presents the result of FST in a dimension of $100 \times 100$, which costs nearly 12 hours for 2000 times Monte Carlo simulations [29] by Qutip [32]. In contrast, there are only 37 clusters in the 4-th order QCE used, and the calculate takes only 1 second on the same computer, showing a speedup over $10^4$ times.

Figure 6(b) shows the quantitative result for the time cost to complete a single simulation task via FST and QCE of different orders. It is clear that the time cost for FST increases exponentially with the pump power while the time costs for QCE approaches are almost constants. Furthermore,
the two approaches scale differently with the system dimension, as shown in Fig. 6(c). For the open system with $m$ modes, the number of equations to solve with FST is $n_{\text{true}}^m / 2$, which increases exponentially with $m$ (dashed lines). Thus, the FST approach becomes impractical when the photon number exceeds 1000 for $m \geq 5$, as implied by the quantum supremacy [33]. Fortunately, for the QCE, the exponential scaling is reduced down to the polynomial relationship between the number of clusters and the number of modes, as shown by the solid lines in Fig. 6(c). To the 2nd-order expansion, the maximum number of items is $f(2)(m)$ tracked in the differential equations is $m^2 + 2m$, which grows quadratically with mode number. For $n$th-order expansion, the number of items is still a polynomial function of mode number with $O(m^n)$. Even though these clusters couple with each other nonlinearly, the computation complexity still follows a polynomial relationship with the number of modes, thus demonstrating the superiority of QCE for quantum many-body physics in multimode bosonic systems.

V. CONCLUSION

In summary, we use the quantum cluster-expansion approach to investigate the classical-to-quantum transition of multimode nonlinear optical systems. The $\chi^{(2)}$ nonlinearity is investigated as a signature two-mode system, with pump laser driving either the fundamental mode or second-harmonic mode. We have discussed the system dynamics with various nonlinear coupling strengths to dissipation ratio $g/\kappa$, and different approaches including the MFA, FST, and the QCE are numerically implemented and compared. It is found that the QCE approach could bridge the gap between the MFA and FST, i.e., capture both the classical behaviors and also the quantum correlations between modes, is appropriate for the nonlinear optical system with a large number of modes and also relatively strong excitations with $g/\kappa \lesssim 1$. QCE greatly reduces the computational complexity and can be applied to describe a wide range of applications based on bosonic oscillators with moderate anharmonicity, including the rapidly-developing integrated nonlinear photonics [15–17, 34] and the superconducting cavity with kinetic inductance [35, 36]. The experimental progress raises urgent needs for the theoretical simulation of nonlinear photonic systems with high efficiency.

Acknowledgments

Y.-X.H. thanks Qianhui Lu and Mingda Li for their supports and inspiring ideas in numerical calculations. This work was funded by the National Key R&D Program (Grant No. 2017YFA0304504), the National Natural Science Foundation of China (Grants No. 11874342, No. 11922411, No. 12061131011, and No. 11904316), Anhui Province Natural Science Foundation (No. 2008085QA34). ML and CLZ were also supported by the Fundamental Research Funds for the Central Universities (Grant Nos. WK2470000031 and WK2030000030), and the State Key Laboratory of Advanced Optical Communication Systems and Networks. The numerical calculations in this paper were partially done on the supercomputing system in the Supercomputing Center of University of Science and Technology of China.
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