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Manifestation of quantum chaos in second-harmonic generation

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ABSTRACT

Second-harmonic generation as a frequency doubling process is ubiquitous in modern optical systems. The present study explores prospects of the stability region of second-harmonic generation in a medium coupled to a bosonic bath. We show that a stability–instability transition due to the combined action of driving field and nonlinearity coupling is seen. We report a reliable evidence confirming the appearance of the chaos in second-harmonic generation under suitable conditions. By tracing direct signatures of chaos in the quantum mechanics, adjacent-spectral-spacing-ratio (ASSR) distribution and participation ratio, we also find a critical point $(\epsilon_c, \kappa_c) = (2.8, 1.1)$ for which a pronounced delocalized regime is seen. To verify the existence of chaotic behavior we also turn our attention on long-range correlation statistics.

1. Introduction

In many cases, the dynamical response of quantum systems is affected by the interaction with an environment, which may be a set of oscillators or a heat bath [1,2]. Thus, a correct investigation of such open quantum systems are based on tracing the effects of interaction with the reservoir, and so is rather complicated from mathematical point of view.

Although the traditional way to describe the dynamics of such systems is the master equation, it conveys some drawback. The main feature of master equation approach is neglecting memory effects. Of course, this assumption does not always hold, e.g., when non-damping or non-oscillating terms are exist. This constraint which may lead to instability of the system, reduces the applicability of the master equation approach [3]. In addition to master equation being a customary approach to solve Markovian processes, some other methods including a non-Markovian quantum jump method [4,5], doubled or tripled Hilbert space methods [6,7], and a non-Markovian quantum state distribution method [8, 9] have also previously been used to simulate non-Markovian processes. Generally, owing to setting different approximations the ability of these methods to apply in general quantum systems are under question [10]. However, an alternative approach is spectral analysis. Besides the ability of spectral analysis to simulate a diversity of dynamical systems [11–15], it can be applied to solve quantum systems interacting with a reservoir without any approximations [10,16].

Generally, the study of quantum optical systems inside a medium coupled a reservoir is of great interest in the fields of nonlinear optics [17–19]. One engaging effect to consider is second-harmonic generation (SHG) [20–22]. The SHG occurs as a result of the atomic response, which causes the intensity of the second-harmonic wave to increase as the square of the intensity of the applied laser light [23]. SHG also known as frequency doubling was first detected and studied in the seminal paper by Franken et al., [24]. Accordingly, the stability of nonlinear optical effects exposed to an incident field is the precursor of phase transition studies. On this matter, the first demonstration of instability and optical chaos in SHG was reported by Savage and Walls [25]. In contrast to most of the previously encountered studies [25–30], which have analyzed the classical signatures of chaos in SHG, surprisingly a reliable evidence confirming the appearance of quantum indicators of chaos in SHG is still lacking. The level repulsion as a principal hallmark of quantum chaos follows from symmetry reduction which mixes states, and gives rise to overlapping of states and so avoided crossings. Under these conditions, the zero energy separation becomes unlikely, and the system may demonstrate level repulsion [31]. We expect the SHG effect be a promising candidate for the observation of level repulsion and so quantum chaos, owing to the inherently reduced symmetry of the crystal lattice, as a result of the incident electric fields [32–34].

In a previous effort [35], we have reported the existence of quantum chaos in the SHG, without considering the effect of bath. Here, we try to extend the observations to a general case

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by considering the case of SHG inside a medium coupled to a bosonic bath. The investigation of such phenomena is possible in the framework of toy-model based on the n-photon states approximations. Concerning this, we try to proceed our aim based on the two-photon states approach. Based on these considerations, we use spectral analysis to explore dynamical response and phase diagram of SHG. We find that the combined action of increasing nonlinearity and driving field intensity induces an integrable-chaotic transition. We address the observed transition theoretically, without discussing the enhancement of SHG.

The remaining of the study is assembled as follows. In Section 2 we review the starting model describing the interaction of light modes with frequencies ω and 2ω. Then, in sections 3, 4 we report our results for a defined dimension of the system. In section 3 we address the onset of electric-field-induced delocalization, with emphasis on the quantum chaos results. Section 4 is aimed at investigating the effect of nonlinearity on the integrable-chaos transition of SHG. Finally, in Section 5 we conclude our observations.

2. Model

The Hamiltonian for the interaction of a light mode at frequency ω with its second harmonic at frequency 2ω in the rotating wave approximation is written as [36,37]

$$H = H_{int} + H_P + H_B, \tag{1}$$

where the interaction term of Hamiltonian in the $\hbar = 1$ basis is

$$H_{int} = i\frac{\kappa}{2}(a^2 b^\dagger - a^{\dagger 2} b), \tag{2}$$

where κ, representing the effective $\chi^{(2)}$ coupling strength between the two modes, stands for nonlinearity strength. a is the bosonic annihilation operator for excitations at frequency ω and b annihilates excitations at frequency 2ω.

By considering the perturbation of the system from the outside by a coherent field with the frequency of the fundamental mode, the pumping term of Hamiltonian is described as

$$H_P = i(\epsilon a^\dagger - \epsilon^* a), \tag{3}$$

where ϵ is the classical amplitude of the pump. The semiclassical approximation assumes that the electric field can be treated classically [38].

Due to the loss of light through the partially transmitting mirrors of the cavity, the system of interest is dissipative. Modeling of this loss is provided by coupling the cavity modes to reservoir as

$$H_B = \Gamma_a^\dagger a + \Gamma_a a^\dagger + \Gamma_b^\dagger b + \Gamma_b b^\dagger. \tag{4}$$

Γ_a and Γ_b are bath operators and represent cavity losses for the two modes.

For further consideration we represent the Hamiltonian in the Fock space matrix form. The Fock space representation of a state vector of the system is expressed as [39]

$$|\Psi^{(n)}(\epsilon,\kappa)> = \sum_{\alpha,\beta,\zeta,\eta}^{N} C^{(n)}_{\alpha\beta\zeta\eta}(\epsilon,\kappa)|\alpha>|\beta>|\zeta>|\eta>, \tag{5}$$

where the index (n) indicates the n-th member of the ensemble of state vectors. $|\alpha>$ and $|\beta>$ are the Fock states of the fundamental and the second-harmonic modes, respectively. $|\zeta>$ and $|\eta>$ do the same for cavity modes. N defines the dimensionality of the system, in a way that dimension of the whole Fock space equals $dim(\mathcal{H}) = N^4$. The power 4 stands for the 4 Fock states, defining the basis of the space we work. The matrix elements of Hamiltonian is expressed as follows

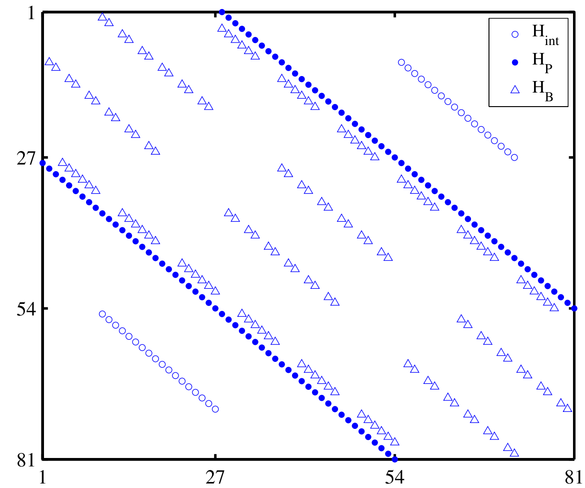

Fig. 1. (Color online.) Schematic representation of non-zero elements of Hamiltonian for $N = 3$.

$$\begin{cases} H^{(int)}_{\alpha'\alpha,\beta'\beta,\zeta'\zeta,\eta'\eta} = i\frac{\kappa}{2}\Big(\sqrt{\alpha(\alpha-1)(\beta+1)}\,\delta_{\alpha',\alpha-2}\,\delta_{\beta',\beta+1} - \sqrt{(\alpha+1)(\alpha+2)\beta}\,\delta_{\alpha',\alpha+2}\,\delta_{\beta',\beta-1}\Big) \times \delta_{\zeta',\zeta}\,\delta_{\eta',\eta}\,, \\ H^{(P)}_{\alpha'\alpha,\beta'\beta,\zeta'\zeta,\eta'\eta} = i\Big(\epsilon\sqrt{\alpha+1}\,\delta_{\alpha',\alpha+1} - \epsilon^\star\sqrt{\alpha}\,\delta_{\alpha',\alpha-1}\Big) \times \delta_{\beta',\beta}\,\delta_{\zeta',\zeta}\,\delta_{\eta',\eta}\,, \\ H^{(B)}_{\alpha'\alpha,\beta'\beta,\zeta'\zeta,\eta'\eta} = \sqrt{\alpha(\zeta+1)}\,\delta_{\alpha',\alpha-1}\,\delta_{\beta',\beta}\,\delta_{\zeta',\zeta+1}\,\delta_{\eta',\eta} + \sqrt{(\alpha+1)\zeta}\,\delta_{\alpha',\alpha+1}\,\delta_{\beta',\beta}\,\delta_{\zeta',\zeta-1}\,\delta_{\eta',\eta} + \sqrt{\beta(\eta+1)}\,\delta_{\alpha',\alpha}\,\delta_{\beta',\beta-1}\,\delta_{\zeta',\zeta}\,\delta_{\eta',\eta+1} + \sqrt{(\beta+1)\eta}\,\delta_{\alpha',\alpha}\,\delta_{\beta',\beta+1}\,\delta_{\zeta',\zeta}\,\delta_{\eta',\eta-1}\,. \end{cases} \tag{6}$$

The schematic picture of the non-zero elements of Hamiltonian (1) for $N = 3$ is shown in Fig. 1. Note that we report our obtained results for $N = 7$ in following sections 3 and 4.

At the considered electric field intensity and the nonlinearity of medium, $\chi^{(2)}$ approximations is applicable [39,40]. Hence, one can explore the dynamical response of the system in the framework of two-photon states [39,40], and three-photon states [41]. Here, according to the applied Hamiltonian we consider two-photon states approach.

3. Electric field effect

Contributions to the even order nonlinear optical effects, and consequently SHG, are usually expected to vanish in the case of nanomaterials having symmetric structure [23,42]. Non-vanishing contributions to such nonlinear optical effects are expected to appear when the symmetry is broken [43,44]. Applying electric fields is one of the commonly used approaches to remove the symmetry and the enhancement of nonlinear effects [32,33]. Besides removing the symmetry, the applied electric field leads to the non-separability of Schrödinger's equation. In this regime, instability and consequently quantum chaotic behavior is expected.

In order to determine the instability region of the studied system subjected to a controllable electric field, we use the spectral analysis approach. Owing to the loss of trajectory notion in quantum mechanics, instability in quantum mechanics cannot be further explored in a similar way with classical physics [45]. According to the Bohigas–Giannoni–Schmit conjecture [46], inspired by the work of Casati et al. [47], the statistical properties of the set of energy levels of a given system in the semiclassical limit [48,49] are well described by random matrix theory. In this regard, different random matrices are mapped to different physical