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Instability in Third-Harmonic Generation

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Abstract. Large optical nonlinearities are critical to photonic technologies, and generation of harmonics is a convenient way of producing new wavelengths of light. In particular, third-harmonic generation from infrared sources is often used as a source of ultraviolet light. In this paper, we study the stability of the dynamics of the process of intracavity generation of third harmonic. We show that owing to a rather general phenomenon analogous to Anderson transition, a stability-instability transition due to the combined action of driving field and nonlinearity coupling is seen. It is shown that the dynamics of the system strongly depends on the external electric field of the fundamental mode and on the coupling coefficient of the interacting modes.

Keywords: Third harmonic generation, Instability, Quantum chaos.

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 the non-Markovian quantum jump method [4,5], doubled or tripled Hilbert space methods [6,7], and the 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

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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 [11,12], it can be applied to solve quantum systems interacting with a reservoir without any approximations [10].

Generally, the study of quantum optical systems interacting with a reservoir is of great interest in the fields of nonlinear optics (NLO) [13].Among the various NLO phenomena the (third-harmonic generation) THG is a commonly used nonlinear optical process for the efficient frequency tripling [14–16] and is of special interest for possible applications in optoelectronic devices and graphene nanotubes [17,18].

To date, most efforts have been devoted to improving the efficiency of the THG. Accordingly, the stability of NLO phenomena exposed to an incident field is the precursor of phase transition studies. The first theoretical analysis of the THG backs to the work of Armstrong *et al.* in 1962 [19]. In this regard, the intracavity second-harmonic generation (SHG) is rather well studied. Studies [20–23] are directed at investigating the dynamical response of the intracavity SHG above the bifurcation point. However, as compared to the intracavity SHG, the intracavity THG process is insufficiently explored.

In present study, we try to extend the observations to a general case by considering the case of interacting THG with a bosonic bath. Based on these considerations, we use spectral analysis to explore dynamical response and phase diagram of THG interacting with a bosonic bath. 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 THG.

2 Model

We study the THG model inside a two-mode cavity in the $\hbar = 1$ basis. To this end, we consider that a nonlinear medium with an appropriate third-order susceptibility $\chi^{(3)}$ is placed inside the cavity tuned to the frequencies of the fundamental ω and of the third harmonic 3ω modes. By considering perturbation of the fundamental mode by an external classical coherent field ϵ, the interaction of a light mode at frequency ω with its third harmonic in the rotating wave approximation is described by the following Hamiltonian [24]

$$H = i\frac{\kappa}{2}(a^3 b^\dagger - {a^\dagger}^3 b) + i(\epsilon a^\dagger - \epsilon^* a) + \Gamma_a^\dagger a + \Gamma_a a^\dagger + \Gamma_b^\dagger b + \Gamma_b b^\dagger, \qquad (1)$$

where κ, representing the effective nonlinear third-order susceptibility $\chi^{(3)}$, holds for coupling coefficient between the two modes. $a(a^\dagger)$ is the bosonic annihilation (creation) operator for excitations at fundamental frequency and $b(b^\dagger)$ do the same at its third harmonic.

The semiclassical approximation assumes that the electric field can be treated classically [25].

Due to the loss of light through the partially transmitting mirrors of the cavity, the system of interest is dissipative. Γ_a and Γ_b are thermostat operators and represent cavity losses for the two modes.

3 Quantum chaos signatures

We introduce aspects of instability and quantum chaos signatures in the interacting modes of the intracavity THG. The notion of classical chaos is reflected in phase-space trajectories. Missing link between the signatures of chaotic systems in classical and quantum domains due to the loss of phase-space trajectory concept in quantum mechanics, is established by Wigner in the framework of random matrix theory (RMT) and in terms of their energy level fluctuations.
The properties of quantum systems with classical chaotic counterparts are different from those with classical regular counterparts. Where the energy levels of a chaotic quantum system are highly correlated, a regular (integrable) system exhibits an uncorrelated feature [26,27]. Hence, RMT characterizes quantum systems by classifying their spectral fluctuations in terms of distinct symmetry classes such as Gaussian orthogonal (GOE) and Poissonian ensembles. Remarkably, the BGS conjecture [28] further enriched the field of RMT, showing that GOE applies generally to chaotic systems in the semiclassical limit [29].
Following RMT, the adjacent-spectral-spacing-ratio (ASSR) distribution $P(r)$ [30] defines the spectral fluctuations of quantum systems by measuring short range correlations between ASSRs defined as $r_n = \frac{s_n}{s_{n-1}}$, where $s_n = E_n - E_{n-1}$ is the space between any two adjacent levels. As long as the average level density does not change too much on the scale of level spacings, the unfolding procedure is not needed for r. For uncorrelated levels, a Poissonian distribution $P(r) = \frac{1}{(1+r)^2}$ with a mean value of $\gamma_P \doteq <\gamma>_{Poisson} = 0.3863$ holds for ASSR distribution. On the other hand, for quantum chaotic systems with linear degree of level repulsion, correlations are strong and the Wigner-Dyson distribution, $P(r) = \frac{27}{8}\frac{r+r^2}{(1+r+r^2)^{(\frac{5}{2})}}$ with $\gamma_{GOE} \doteq <\gamma>_{GOE} = 0.5359$ stands [30]. Note that the set of γs is defined as $\gamma_n = \min(r_n, \frac{1}{r_n})$.
Due to the fact that higher-order nonlinearities couple envelopes with different carrier frequencies, it is expected that an additional complexity arises [31]. Applied field leads to the non-separability of the Schrödinger's equation. Furthermore, previous studies on the dynamics of the number of photons show that in the case of nonlinear optical processes, such as, e.g., the intracavity SHG and THG [32,33], stationary solutions are stable only for relatively small perturbations. For these systems, a critical pump field was reported above which small fluctuations in the system do not decay and self-sustained oscillations in the photon number dynamics was observed. As a result, it seems that increasing the external resonant perturbation to be followed with the break down of integrability in the domain of proper nonlinearities. As is expected, the combination increase of the electric field intensity and the nonlinearity follows with a strong trend for states to mixing. Since this mixing is the main reason for quantum delocalization [27], a transition to chaotic behavior is expected to occur. This expected transition can be seen from Figs. 1,2.
We evaluate spectral fluctuations across the THG spectrum and compare re-

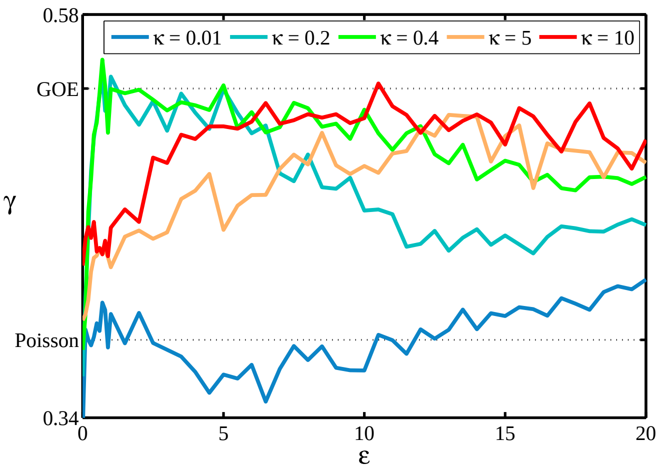

Fig. 1. Dependence of γ on the electric field.

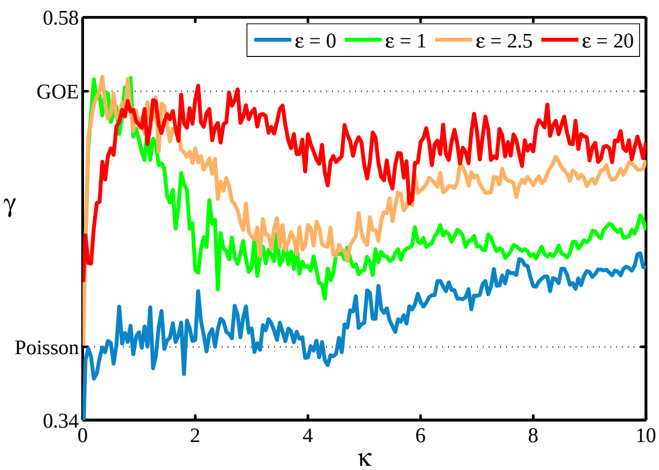

Fig. 2. Dependence of γ on the nonlinearity.

sults to predictions for Poisson and GOE ensembles in Figs. 1,2, where the variation of γ vs. ϵ and κ is depicted. As is seen, for weak ϵs close to 0, results match the prediction for the Poisson ensemble well, even for the larger κs. This result is clearly observable from the Figs. 1,2. A similar behavior is seen for weak κs even for larger fields. As a result, localization of the light in the beginning of dynamics follows from the weakness of either the nonlinearity or the applied field.
Another interesting aspect on the light propagation may be found by looking at the localization properties of the light in the THG process as a function of coherent driving and nonlinearity coupling. We now try to reveal the trace of the observed transition on the structure of the eigenstates. Particularly,

we investigate how their localization characters vary in both regimes of Poissonian and chaotic. To this end, we turn our attention at their components addressing the spreading of the eigenstates. Let consider a typical eigenstate $|\Psi^{(n)}(\epsilon,\kappa)\ >= \sum_{\alpha,\beta,\zeta,\eta}^{N} C_{qp}^{(n)}(\epsilon,\kappa)|\alpha\ >|\beta\ >|\zeta\ >|\eta\ >$ of the Hamiltonian (Eq.(1)). With this consideration, we can define the localization measure of the given state vector as $P^{(n)}(\epsilon,\kappa) = \frac{1}{\sum_{\alpha,\beta,\zeta,\eta}|C_{qp}^{(n)}(\epsilon,\kappa)|^4}$ [34] named as participation ratio. Indeed, the participation ratio $P^{(n)}$, measuring degree of localization [35], gives the number of basis vectors contributing to each eigenstate. It is noteworthy that the participation ratio provides a way of comparison. In particular, it reorganizes itself from being close to 1 for a localized state and approaching $dim(\mathcal{H})(= 2401$ for this study), for a fully delocalized state. It is interesting to look at the ensemble average of the estimated participation ratio. It can be written as $P(\epsilon,\kappa) = \frac{1}{dim(\mathcal{H})}\sum_n P^{(n)}(\epsilon,\kappa)$ [36]. Because the weights $|c_{qp}|^2$ fluctuate, the average over the ensemble gives the number of principal components $\sim \frac{N^2}{3}$ [37,38].

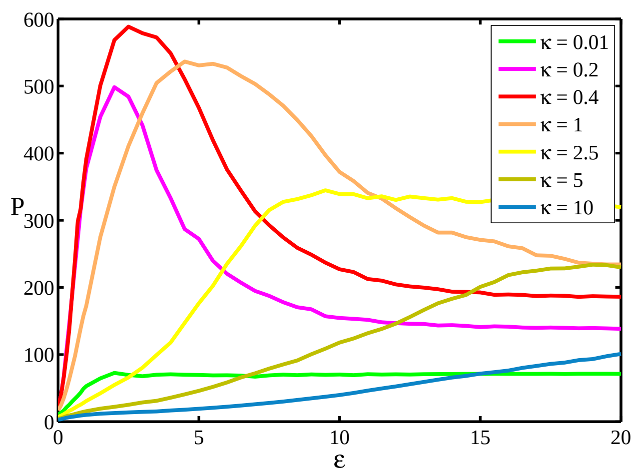

Fig. 3. Dependence of participation ratio on the electric field.

Figures 3,4 shows the variation of P as a function of ϵ and κ. We observe that the states at the edge of weak fields are more localized. Increasing the electric field leads to a clear deviation from localized behavior and a significant increase in the level of delocalization. Especially, in contrast to weak ϵs, the largest values of P are restricted to the middle region of the studied interval. Our numerical computation reveals that the maximum value of P takes place for $(\epsilon_c,\kappa_c) = (3.1, 0.35)$. We refer to this point as the critical point. From the central part of Figs. 3,4 we estimate a typical P of ≈ 700 for (κ_c,ϵ_c), whose order of magnitude indicates strong delocalization. It means that a larger fraction of states are delocalized and widely contribute in the particle transport. This property is a direct consequence of overlapping which leads to widely dis-

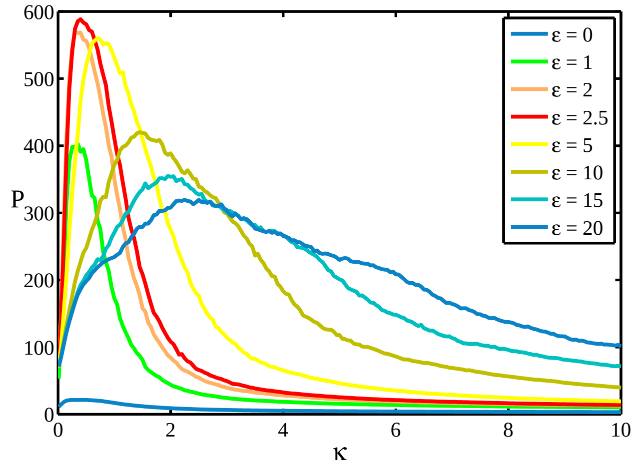

Fig. 4. Dependence of participation ratio on the nonlinearity.

tributed eigenstates. So, the emergence of quantum chaos behavior for this regime seems to be more likely.

4 Conclusion

The generation of the third and high harmonics has been one of the most intriguing phenomena in nonlinear optics. THG is a nonlinear optical phenomenon capable of tripling the efficient frequency over a broad range of wavelengths. Up to now, the effect of the nonlinear loss on the THG efficiency have been studied in detail. In this regard, by deriving the Langevin equations for stochastic field amplitudes for the THG phenomenon and finding its bifurcation point, Gevorkyan *et al.* [33] have shown that above this point, the dynamics of the number of photons of interacting modes changes to the regime of self-sustained oscillations. Moreover, they have reported the appearance of the instability domain beyond the critical point. However, less attention has been paid to the role of the external field and the nonlinearity coupling in driving THG to produce quantum chaotic behavior so far. Therefore, we was motivated to perform present study. Owing to the fundamental role of nonlinear optical materials with large third-order nonlinear susceptibilities we have performed our analyses in the case of weak and strong κs.
Here, we presented a quantum chaos study of intracavity THG. A coherent external classic field was included into the consideration and the corresponding Hamiltonian was expanded in the matrix form. The semiclassical analysis of eigenvalues and the spatial distributions of eigenvectors were performed. It was shown that the THG undergoes a stable-unstable transition driven by the combination action of the electric field and nonlinearity of the medium. Compared with the previous studies exploring Anderson localization in the presence of defects and disorder in a crystal [35], our study provides another approach to-

wards Poisson-GOE transition in THG without introducing on- or off-diagonal disorder. We found that both for weak enough nonlinearity and low intensities of the electric field, the localization phase and steady-state regime dominate. We found that, the steady-state notion in the THG process losses its mean for large enough values of ϵ, κ. Accordingly, for sufficiently strong driving field and nonlinearity a transition to instability and chaotic regime has also been observed. Moreover, we explored delocalization transition through the fluctuation measures of the levels and the state vectors, and we observed that there is a solid evidence for the presence of quantum chaos in suitable values of ϵ, κ. In comparison with studies reporting the appearance of instability and chaos in the THG [32,33], present study benefits from detecting a critical point $(\epsilon_c, \kappa_c) = (3.1, 0.35)$ for which the participation ratio, and so the level of delocalization is maximized. The presence of this critical point foretells a large fraction of delocalized states in comparison with localized ones. It should be noted that in the current study, only the appearance of the critical point has been shown. Furthermore, to approve the occurrence of quantum chaos we concluded that as expected the observed ASSR distribution shows a good harmony with the GOE prediction for the critical point.

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