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# Nonlinear optical dynamics of a 2D semiconductor quantum dot super-crystal: Emerging multistability, self-oscillations and chaos

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**Abstract.** We conduct a theoretical study of the nonlinear optical dynamics of a 2D super-crystal comprising regularly spaced identical semiconductor quantum dots (SQDs), subjected to a resonant continuous wave excitation. A single SQD is considered as three-level ladder-like systems involving the ground, one-exciton and bi-exciton states. We show that the super-crystal reveals a rich nonlinear dynamics, exhibiting multistability, self-oscillations and chaos. The behaviour is driven by the retarded SQD-SQD interactions and bi-exciton binding energy.

## 1. Introduction

In the last decade, the so-called *metamaterials*, a class of new materials not existing in nature, received a great deal of attention [1, 2]). Super-crystals comprising regularly spaced identical SQDs represent one of examples of metamaterials with tunable optical properties which can be controlled by the geometry and chemical composition of components, as well as the lattice arrangement. There exists a variety of methods to fabricate such systems [3, 4].

In this communication, we report on a theoretical study of the nonlinear optical dynamics of a super-crystal comprising a square lattice of spherical SQDs (preliminary results have been published in Ref. [5]). A single SQD is considered as a point-dipole system, that is accurate if the variation of the optical field over the particle size  $d$  is small ( $kd \ll 1$ ,  $k$  being the wavenumber) and  $a > (3/2)d$ , where  $a$  is the lattice constant [6]. We assume that these conditions hold. Due to the high density of SQDs and high oscillator strengths of the SQD's transitions, the total (retarded) dipole-dipole SQD-SQD interactions have to be taken into account, which is elaborated in the paper within the mean-field approximation. The real part of the SQD-SQD interaction results in the dynamic shift of the SQD's energy levels, whereas the imaginary part describes the collective radiative decay of SQDs, both depending on the population differences between the levels. These two effects are crucial for the nonlinear dynamics of the SQD super-crystal. As a result, in addition to bistability, analogous to that for a thin layer of two-level



emitters [8], we found multistability, periodic and aperiodic self-oscillations and chaotic regimes in the super-crystal optical response.

## 2. Theoretical background

We model single SQD by a three-level ladder-like system with the ground, one-exciton, and bi-exciton states ( $|1\rangle$ ,  $|2\rangle$ , and  $|3\rangle$ , respectively) with corresponding energies  $\varepsilon_1 = 0$ ,  $\varepsilon_2 = \hbar\omega_2$ , and  $\varepsilon_3 = \hbar\omega_3 = \hbar(2\omega_2 - \Delta_B)$ , where  $\hbar\Delta_B$  is bi-exciton binding energy (see, e.g., Refs. [7]). The dipole allowed transitions  $|1\rangle \leftrightarrow |2\rangle$  and  $|2\rangle \leftrightarrow |3\rangle$  are characterized by the transition dipole moments  $\mathbf{d}_{21}$  and  $\mathbf{d}_{32}$ , the latter are assumed to be real, aligned, and  $\mathbf{d}_{32} = \mu\mathbf{d}_{21} \equiv \mu\mathbf{d}$ . The states  $|3\rangle$  and  $|2\rangle$  spontaneously decay with rates  $\gamma_{32}$  and  $\gamma_{21}$ , accordingly, and  $\gamma_{32} = \mu^2\gamma_{21} \equiv \mu^2\gamma$ . The bi-exciton state  $|3\rangle$  is dipole forbidden and can be populated either via consecutive  $|1\rangle \rightarrow |2\rangle \rightarrow |3\rangle$  transitions or via the simultaneous absorption of two photons of frequency  $\omega_3/2$ . In what follows, we will consider both options.

It is assumed that the system is subjected to a resonant external field  $\mathbf{E}_0 \cos(\omega t)$  incident normally to the super-crystal and polarized along the transition dipole moments. The optical dynamics of SQDs is described by means of the quantum master equation for the density operator  $\rho$ , which in the rotating (with the frequency of the external field  $\omega$ ) frame reads

$$\dot{\rho} = -\frac{i}{\hbar} [H^{RWA}, \rho] + \frac{\gamma_{21}}{2} ([\sigma_{12}\rho, \sigma_{21}] + [\sigma_{12}, \rho\sigma_{21}]) + \frac{\gamma_{32}}{2} ([\sigma_{23}\rho, \sigma_{32}] + [\sigma_{23}, \rho\sigma_{32}]), \quad (1a)$$

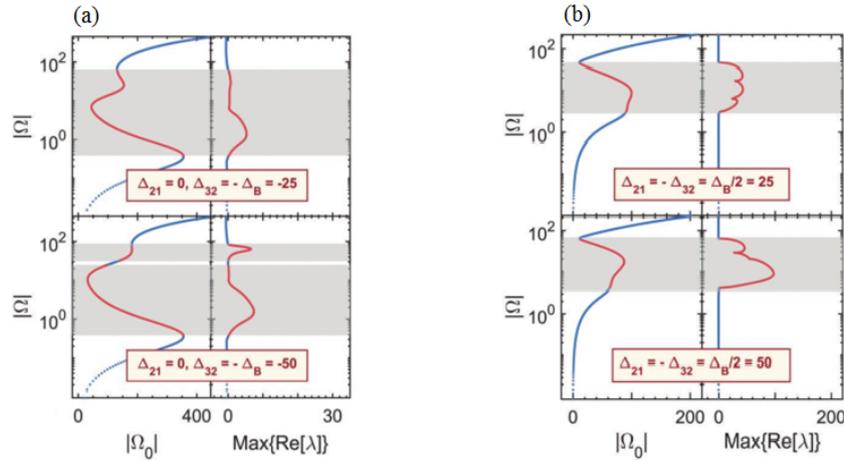
$$H^{RWA} = \hbar [\Delta_{21}\sigma_{22} + (\Delta_{21} + \Delta_{32})\sigma_{33}] - i\hbar\Omega (\sigma_{21} + \mu\sigma_{32}) + h.c., \quad (1b)$$

$$\Omega = \Omega_0 + (\gamma_R + i\Delta_L)(\rho_{21} + \mu\rho_{32}). \quad (1c)$$

In Eq. (1a),  $\hbar$  is the reduced Plank constant,  $H^{RWA}$  is the SQD Hamiltonian in the rotating wave approximation (RWA), square brackets denote the commutator, and the two other terms represent the relaxation operator, where  $\sigma_{ij} = |i\rangle\langle j|$  ( $i, j = 1, 2, 3$ ). In Eq. (1b),  $\Delta_{21} = \omega_2 - \omega$  and  $\Delta_{32} = \omega_3 - \omega_2 - \omega$  are detuning away from the resonance of the corresponding transitions.  $\Omega_0 = dE_0/\hbar$  and  $\Omega = dE_{SQD}/\hbar$  are the Rabi amplitudes of the external ( $E_0$ ) and total field ( $E_{SQD}$ ), acting on a SQD. The latter is the sum of the applied field  $E_0$  and the field produced by all others SQDs in place of a given one [the second term in Eq. (1c)], taken in the mean-field approximation. The quantities  $\gamma_R$  and  $\Delta_L$  are the amplitudes of the far-zone and near-zone fields, describing the collective SQD relaxation and energy level shifts, respectively. They are given by the following expressions [9]:  $\gamma_R = 4.51\gamma/(ka)^2$  and  $\Delta_L = -3.35\gamma/(ka)^3$ .

## 3. Results

In calculations, we set  $\mu = \sqrt{2/3}$ ,  $\gamma_R = 100$ ,  $\Delta_L = 1000$  [9] (here and in the following, all quantities are in units of  $\gamma$ ). The results of calculations are presented in Figs. 1 and 2. Figure 1 displays the steady-state solutions to Eqs. (1a) - (1c) obtained by the method invented in Ref. [9]. Plots (a) and (b) show the dependence of the mean-field magnitude  $|\Omega|$  (left column) on the external field magnitude  $|\Omega_0|$  calculated for the one-photon ( $\omega_0 = \omega_2$ ,  $\Delta_{21} = 0$ ,  $\Delta_{32} = -\Delta_B$ ) and two-photon ( $\omega_0 = \omega_3/2$ ,  $\Delta_{21} = -\Delta_{32} = \Delta_B/2$  resonance, respectively). As is seen from the figures, the mean-field magnitude  $|\Omega|$  may have several solutions (up to five at  $\Delta_B = -25$ ) for a given value of the external field magnitude  $|\Omega_0|$ , that signals emerging instabilities. We uncovered the stability of different branches computing the Lyapunov exponents  $\lambda$  [10]. For this purpose, we calculated the eigenvalues of the Jacoby matrix (of rank eight) of the linearized Eqs. (1a) - (1c) as a function of  $|\Omega|$  and selected from those an exponent with the maximal real part,  $\text{Max}\{\text{Re}[\lambda]\}$ , which determines the character of evolution of a small deviation from the steady-state solution. At  $\text{Max}\{\text{Re}[\lambda]\} < 0$ , a given solution is stable (deviation decreases) and *vice versa*. Calculated in this way  $\text{Max}\{\text{Re}[\lambda]\}$  are plotted in the right panels of Fig. 1. The



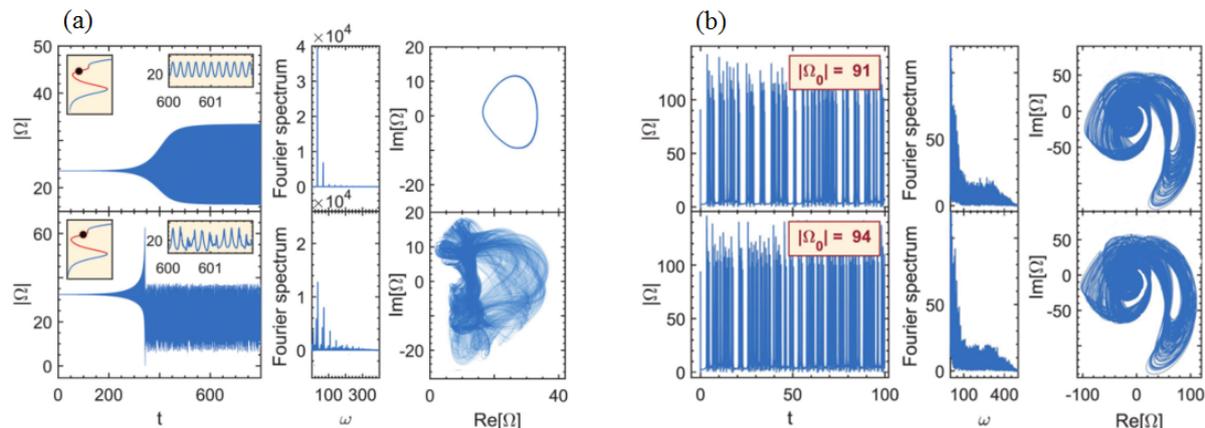
**Figure 1.** (a) - Steady-state solutions to Eqs. (1a) - (1c) for the case of one-photon resonance ( $\Delta_{21} = 0, \Delta_{32} = -\Delta_B$ ) for different values of the bi-exciton binding energy  $\Delta_B$  (shown in the plots). The left column displays the  $|\Omega|$ -vs- $|\Omega_0|$  dependence. The shaded regions show unstable parts of the stationary solutions (marked in red), obtained by analyzing the Lyapunov exponents  $\lambda$ , the maximum values of the real parts of which,  $\text{Max}\{\text{Re}[\lambda]\}$ , are depicted in the right column. (b) - Same as in (a), only for the case of two-photon resonance ( $\Delta_{21} = -\Delta_{32} = -\Delta_B/2$ .)

shaded regions show the unstable ( $\text{Max}\{\text{Re}[\lambda]\} > 0$ ) parts of the steady-state solutions. We stress that not only branches with the negative slope are unstable, that is always the case, but those with the positive slope as well. This occurs for both the one- and two-photon resonance excitation, in the upper branch for the former and in the lower one for the latter.

To uncover the nature of instabilities of different branches of the steady-state solutions (shown in Fig. 1), we solved numerically Eqs. (1a) - (1c). Two types of initial conditions were used: (i) - the system starts from one of the steady-states solutions and (i) - the system initially is in the ground state [ $\rho_{11}(0) = 1$ ]. Figure 2 shows the dynamics of the mean-field  $\Omega$  calculated for the one-photon resonance ( $\omega_0 = \omega_2, \Delta_{21} = 0, \Delta_{32} = -\Delta_B$ ) - plot (a) and two-photon resonance ( $\omega_0 = \omega_3/2, \Delta_{21} = -\Delta_{32} = \Delta_B/2$ ) - plot (b).

Figure 2(a) displays the results obtained for  $\Delta_B = 50$ . Two unstable points of the the steady-state solution were considered: ( $|\Omega_0| = 90, |\Omega| = 23.5655$ ) - upper row and ( $|\Omega_0| = 140.0002$  - lower row (see the inserts). The left column in panel (a) shows the time evolution of the mean-field magnitude  $|\Omega|$ . As is seen, after some delay, the system reaches an attractor (sustained signal), the character of which significantly differs for considered points. The upper row demonstrates results for ( $|\Omega_0| = 90, |\Omega| = 23.5655$ ); the dynamics looks like simple self-oscillations (see the upper-right inset for a blow up of the dynamics of  $|\Omega|$ ). The Fourier spectrum of the sustained signal (middle column) contains a couple of well-defined harmonics. Accordingly, the phase map in the phase space ( $\text{Re}[\Omega], \text{Im}[\Omega]$ ) (right panel) represents a simple closed curve, commonly called limit cycle [10]. By contrast, for ( $|\Omega_0| = 140.0002, |\Omega| = 32.3261$ ), the dynamics manifests signatures of aperiodic oscillations. This is reflected in the Fourier spectrum of the attractor which consists of incommensurated frequencies [10]. The phase space map of the attractor represents an open trajectory, almost filling a finite area in the phase space.

Figure 2(b) shows the evolution of the system, being initially in the ground state [ $\rho_{11}(0) = 1$ ], calculated for the external field magnitudes  $|\Omega_0| = 91$  (upper row) and  $|\Omega_0| = 94$  (lower row) at  $\Delta_B = 50$ . Here, in both cases we observe a highly irregular behavior of the system. The Fourier spectrum of the signal represents a broad structureless quasi-continuum, suggesting



**Figure 2.** (a) - Time-domain dynamics of the mean-field magnitude  $|\Omega|$  (left column), the Fourier spectrum (middle column) and phase space map of the attractor in the  $(\text{Re}[\Omega], \text{Im}[\Omega])$  plane (right column) for the case of the one-photon resonance ( $\Delta_{21} = 0$ ,  $\Delta_{32} = -\Delta_B = -50$ ). The inserts show fragments of the dynamics on a short time interval (upper-right insets) and the steady-state solution (upper-left insets), where the thick dots indicate the points on the steady-state solutions: ( $|\Omega_0| = 90$ ,  $|\Omega| = 23.5655$ ) - upper row, ( $|\Omega_0| = 140.0002$ ,  $|\Omega| = 32.3261$ ) - lower row, for which the calculations were performed. (b) - Same as in (a), but now for the case of the two-photon resonance ( $\Delta_{21} = -\Delta_{32} = -\Delta_B/2 = -25$ ) for the external field magnitudes  $|\Omega_0| = 91$  (upper row) and  $|\Omega_0| = 94$  (lower row) and the ground state as the initial condition.

that the oscillatory regime found is of chaotic nature [10]. The phase-space map shows the corresponding attractor. In the chaotic regime, the system behavior is very sensitive to the initial conditions, that may be used for the encryption of information [11].

#### 4. Summary

We conducted a theoretical study of the nonlinear optical dynamics of a 2D SQD super-crystal subject to a resonant continuous wave excitation, modeling an isolated SQD a three-level ladder-like system with the ground, one-exciton and biexciton states. The set of parameters used in our study is typical for SQDs emitting in the visible range, such as, for instance, CdSe and CdSe/ZnSe. It turned out that the super-crystal optical response might demonstrate multistability, self-oscillations and dynamic chaos. All this makes such systems promising for practical applications in all-optical information processing and optical computing.

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