Nonlinear optical dynamics of two-dimensional super-lattices of quantum V-emitters

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Abstract. We study theoretically the nonlinear optical response of a super-lattice of regularly arranged three-level identical quantum emitters with a doublet in the excited state to the action of a monochromatic electromagnetic field quasi-resonant to optical transitions in the emitter, using into account the dephasing of the system. The total retarded dipole-dipole interaction of the emitters is accounted for in the mean-field approximation. This interaction plays the role of positive feedback, which (in combination with the immanent nonlinearity of emitters themselves) leads to multistability of the super-lattice response. The stability of different response branches is analyzed using the Lyapunov exponents' method. Another important property of the super-lattice is its high reflectance in a certain frequency range; i.e., within this range, the super-lattice operates as a perfect nanometer mirror; moreover, reflection can be switched to transmission changing slightly the incident field amplitude (bistability). The possibility of the application of the above-mentioned super-lattice optical properties in nanophotonics is discussed.

1. Introduction

The methods of modern micro-and nanotechnologies make it possible to obtain objects with unusual electromagnetic properties (so-called metamaterials [1, 2, 3]), among which 2D supercrystals (SCs) of semiconducting quantum dots (QDs) [4, 5, 6] and organic polymers [7] are of special interest. The SC optical properties depend on the size of QDs, their shape, chemical composition, lattice geometry and can be purposefully controlled (see [8] and the literature cited therein).

In recent publications [9, 10, 11, 12] was shown that the nonlinear response of a SC of quantum emitters (QEs) with a ladder scheme [9, 10, 11] and Λ, V scheme of optical transitions [12, 13, 14] exhibits multistability, self-oscillations, and dynamic chaos. Also, in a certain frequency band, the SC almost completely reflects the incident field, i.e., it is an ideal nanometer mirror, which, moreover, at certain frequency characteristics of the emitters can be bistable, even under the conditions of dephasing the system.

In this study, we analyze theoretically the nonlinear optical response of super-lattice consisting of regularly spaced QEs with a doublet in the excited state (V-QEs) under the condition of dephasing the system. The role of such an emitter can be played, for example, by semiconductor
QDs with a degenerate valence band in a magnetic field \[15\]. Because of the high density of V-QEs and their large oscillator strength of transitions, the dipole–dipole interaction of V-QEs plays a crucial role in the optical response of the super-lattice. Since the quantum-mechanical mean dipole moment of a V-QE depends on the current quantum state of the latter, the QE–QE interaction is also a function of this state. This ensures the positive feedback which, in combination with the immanent nonlinearity of the V-QE itself, leads to multistability of the QE–QE interaction.

2. Model

We consider a super-lattice of regularly spaced identical V-QEs under the condition of dephasing the system. The diagram of energy levels and transitions in an isolated V-QE is represented in the following form - the lower ground state |1\> with energy \(\varepsilon_1 = \hbar \omega_1 = 0\), the excited upper state |2\> and |3\>, a doublet with energies \(\varepsilon_3 = \hbar \omega_3\), and \(\varepsilon_2 = \hbar \omega_2\), respectively. Optically allowed transitions are |1\> ↔ |2\> and |1\> ↔ |3\>, which are characterized by the transition dipole moments \(\mathbf{d}_{21}\) and \(\mathbf{d}_{31}\) (we assume for simplicity that both are real-valued and have the same direction). The states |2\> and |3\> of the doublet decay spontaneously to the ground state |1\> with rates \(\gamma_{21}\) and \(\gamma_{31}\), respectively. Nonradiative relaxation in the doublet is taken into account by constant \(\gamma_{32}\). And dephasing of the system is characterized by the dephasing rate \(\Gamma\).

The super-lattice undergoes a quasi-resonant continuous wave external field \(E = \bar{E}_0 \cos(\omega_0 t)\) of amplitude \(\bar{E}_0\) and frequency \(\omega_0\) incident normally to the super-lattice and polarized along the transition dipole moments. The optical dynamics of V-QEs in a super-lattice is governed by the the system of equations for the density matrix \(\rho_{\alpha\beta}\) \((\alpha, \beta = 1, 2, 3)\), which within the rotating wave approximation reads:

\[
\dot{\rho}_{11} = \gamma_{21} \rho_{22} + \gamma_{31} \rho_{33} + \Omega^* \rho_{31} + \Omega \rho_{31}^* + \mu(\Omega^* \rho_{21} + \Omega \rho_{21}^*) ,
\]

\[
\dot{\rho}_{22} = \gamma_{32} \rho_{33} - \gamma_{21} \rho_{22} - \mu(\Omega^* \rho_{21} + \Omega \rho_{21}^*) ,
\]

\[
\dot{\rho}_{33} = -\gamma_{31} \rho_{31} - \gamma_{21} \rho_{22} - \gamma_{32} \rho_{33} - \Omega^* \rho_{31} \rho_{31} - \Omega \rho_{31}^* \rho_{31}^* ,
\]

\[
\dot{\rho}_{31} = -i(\Delta_{31} + (\gamma_{31} + \gamma_{32})/2 + \Gamma) \rho_{31} + \Omega(\rho_{33} - \rho_{11}) + \mu \rho_{32} ,
\]

\[
\dot{\rho}_{21} = -i(\Delta_{21} + (\gamma_{21} + \gamma_{22})/2 + \Gamma) \rho_{21} + \mu \Omega(\rho_{22} - \rho_{11}) + \Omega \rho_{32}^* ,
\]

\[
\dot{\rho}_{32} = -i(\Delta_{32} + (\gamma_{31} + \gamma_{32})/2 + \Gamma) \rho_{32} - \mu \Omega^* \rho_{33} - \Omega \rho_{21}^* ,
\]

\[
\Omega = \Omega_0 + (\gamma_R - i \Delta_L)(\rho_{31} + \mu \rho_{32}) .
\]

where \(\Delta_{31} = \omega_0 - \omega_{31}\) and \(\Delta_{32} = \omega_0 - \omega_{32}\) are detunings of the incident field frequency \(\omega_0\) from the resonance frequencies \(\omega_{31}\) and \(\omega_{32}\) of the |1\> ↔ |3\> and |2\> ↔ |3\> transitions, respectively, and \(\Gamma\) is the dephasing rate (for the sake of simplicity, the same for both transitions). Furthermore, where \(\Omega_0 = d_{31} \bar{E}/\hbar\), given by Eqs. (7), is the Rabi amplitude of the mean field with \(E\) being the amplitude of the latter, \(\hbar\) is the reduced Plank constant, \(\Omega_0 = d_{31} \bar{E}/\hbar\) stands for the Rabi amplitude of the incident field. The second term in Eqs. (7) represents the Rabi amplitude of the mean field produced by all other emitters at the position of a given one. A part proportional to \(\gamma_R\) describes the far-zone contribution to the mean field, while the one scaled with \(\Delta_L\) accounts for the near zone part which is analogous to the Lorentz local field.

The parameter \(\Delta_L\) is practically independent of the system size; it is nothing but the near-zone dipole-dipole interaction of a given V-emitter with all others. It determines the (excitonic) renormalization of the energy level. Note that at \((X' \gg a)\), regardless of the grating size, the relation \(\Delta_L \gg \gamma_R\), is satisfied, which is decisive for the optical dynamics of the super-lattice.
3. Results

We performed numerical calculations of the optical response of the super-lattice, fixing the constants that determine $\gamma_R$ and $\Delta_L$ similar to two-dimensional super-lattices of semiconductor V-QEs [11, 13]: $\lambda = 100 - 200$ nm, $a = 10 - 20$ nm and $\gamma_{31} = 3 \cdot 10^9$ s$^{-1}$. Then the typical values of the parameters are $\gamma_R = 100 \gamma_{31}$ and $\Delta_L = 1000 \gamma_{31}$. The relaxation constant in the $\gamma_{32}$ doublet was chosen as given, $\gamma_{32} = 0.01 \gamma_{31}$. All calculations were performed under the assumption $\gamma_{21} = \gamma_{31}$ ($\mu = 1$). In what follows, all values of the dimension of frequency are given in units of $\gamma_{31}$, and time - in units of $\gamma_{31}^{-1}$.

![Figure 1](image)

**Figure 1.** The steady-state solution, dynamics, spectrum, and phase diagram of the optical response when the system is excited at the center of the doublet of a quantum V-emitter ($\Delta_{31} = \Delta_{32}/2$) in the presence of dephasing of the states of the emitter: a) is the steady-state solution $|\Omega|$ ($|\Omega_0|$) of the system of equations (1-7), b) is the leading Lyapunov exponent max Re[$\Lambda$], solid (dashed) parts of the curves correspond to stable (unstable) parts of stationary solutions, c,d) - dynamics of super-lattice radiation and the Fourier spectrum of the attractor, e) - the phase portrait of the attractor $\omega$ on the plane (Re[$\Omega$], Im[$\Omega$]).

When a system of emitters is excited to the center of a doublet of an isolated quantum V emitter in the presence of phase relaxation in a stationary solution, the instability of the system is realized in a narrow range of input field values $\Omega_0 \in [35.5, 37.3]$ (fig.1).

For a super-lattice of a system of quantum emitters, the influence of the dipole-dipole interaction $\Delta_L$ leads to a shift of energy levels, as a result of which the resonance level is shifted by $\Delta_{31} = 2\Delta_L$. In fig.2 presents analytical solutions of the nonlinear reflection coefficient $R$ for various values of phase relaxation in the vicinity of the real resonance, which is taken into account by the detuning $\Delta_{31}$.

4. Summary

We carried out a theoretical study of the optical response of a two-dimensional super-lattice of regularly spaced quantum emitters with a doublet in an excited state (type V), taking into account their total retarded dipole-dipole interaction, which was considered in the mean-field approximation with allowance for phase relaxation.

This interaction, due to its dependence on the current quantum state of the emitter, provides positive feedback, which, together with the immanent non-linearity of the emitters themselves, leads to bistability, periodic self-oscillations. Phase relaxation leads to suppression of the reflectivity of the super-lattice. Supercrystals of semiconductor V-QEs with a degenerate valence band in a magnetic field [15], which causes the Zeeman splitting of the conduction band of a quantum dot, can be considered as an implementation of such a system. Such systems appear to be promising for applications in nano-photonics.
**Figure 2.** Nonlinear coefficient of reflection $R$ of the super-lattice on the intensity at different values of detuning $\Delta_{31}$ from resonance and the magnitude of phase relaxation. Doublet splitting $\Delta_{32}$ is shown above the panels. Solid (dashed) portions of the curves correspond to stable (unstable) regions of $R$.

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