A monolayer of three-level quantum $\Lambda$-emitters: A perspective system from the viewpoint of nonlinear optical dynamics and nanophotonics

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Abstract. The nonlinear optical response of a monolayer of regularly spaced three-level quantum emitters with a doublet in the ground state is studied theoretically. It is found that the system demonstrates multistability, self-oscillations and dynamical chaos. In a certain frequency range, the monolayer operates as a perfect bistable mirror.

We perform a theoretical study of the optical response of a monolayer of regularly spaced three-level quantum emitters (QEs) with a doublet in the ground state ($\Lambda$-type QE). The total (retarded) dipole-dipole interaction of QEs is taken into account. This interaction provides a positive feedback which, interplaying with the immanent QE’s nonlinearity, results in multistability, self-oscillations and dynamical chaos of the response. In a certain frequency range, the monolayer operates as a perfect nanometric bistable reflector.

The monolayer is assumed to undergo an action of a CW external field of a Rabi amplitude $\Omega_0$ and frequency $\omega_0$, which is quasi-resonant with the QE’s allowed transitions. A QE is modelled as a three-level $\Lambda$-type quantum system with the excited state $|3\rangle$, and a doublet $|1\rangle$ and $|2\rangle$ in the ground states. Only the optical transitions $|1\rangle\leftrightarrow|3\rangle$ and $|2\rangle\leftrightarrow|3\rangle$ are dipole-allowed. They are characterized by the transition frequencies $\omega_{31}$ and $\omega_{32}$, and spontaneous decay constants $\gamma_{31}$ and $\gamma_{32}$. The relaxation within the doublet with the splitting $\Delta_{21}$ is described by the constant $\gamma_{21}$.

The optical dynamics of a QE is governed by the 3x3 density matrix. The field $\Omega$, acting on a given QE in the monolayer, represents a sum of the external field $\Omega_0$ and the field produced by all other QEs in place of the given one. The near-zone (far-zone) part of the QE-QE interaction gives rise to a dynamic shift of the transition frequencies $\omega_{21}$ and $\omega_{32}$ (collective radiative relaxation of QEs), depending on the population difference of corresponding transitions [1,2], governing by the constants $\Delta_1$ (shift) and $\gamma_R$ (relaxation).

In Fig. 1, we present the result of calculations performed for the case when the external field is tuned into the resonance with the transition $|1\rangle\leftrightarrow|3\rangle$ ($\Delta_{31} = \omega_{31} - \omega_0 = 0$). Panel (b) shows the steady-state solution $|\Omega|\text{vs}.|\Omega_0|$ for the set of parameters [2,3] given in the figure.

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Fig. 1. (a) – Scheme of QE excitation. (b) – The monolayer’s steady-state response. (c) – The real part of the major Lyapunov exponent. Solid (dashed) parts indicates stable (unstable) of the solution. (d), (e) and (f) – Respectively, dynamics, Fourier spectra and phase trajectories of the system calculated for the field magnitudes $|\Omega_0|$ shown in panel (d). Parameters of calculations are: $\Delta_21 = 100$, $\gamma_R = 100$, $\Delta_L = 1000$, $\gamma_{21} = 0.01$. All quantities are given in units of $\gamma_{31}$.

As is seen, $|\Omega|$ turns out to be a multi-valued function of $|\Omega_0|$. The stability of different parts of the solution has been explored analyzing the Lyapunov’s exponents $\Lambda_k$ ($k = 1,2…8$). The maximal real part of $\{\Lambda_k\}$, $\text{Max}\{\text{Re}[\Lambda]\}$, is plotted in panel (c). Plots (d), (e) and (f) display, respectively, dynamics, Fourier spectra and phase maps of the signals obtained for two values of $|\Omega_0|$ shown in the plots. For $|\Omega_0| = 200\gamma_{31}$, the dynamics demonstrates self-oscillations, while for $|\Omega_0| = 302.4\gamma_{31}$, (quasi)chaotic behaviour.

Fig. 2 shows the power refleciton coefficient $R$ calculated in the vicinity of the resonance renormalized by the QE-QE dipole-dipole interaction, $\Delta_{31} = \Delta_L = 1000\gamma_{31}$ [1,2]. As follows from the figure, the monolayer exhibits high reflectance and bistability [panel (b)] as well as self-oscillations [panel (c)].

Summarizing, a monolayer of $\Lambda$-type QEs demonstrates multistability, self-oscillations, dynamical chaos and high reflectance, the features being perspective for nanophotonics.

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References