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Interaction of ultrashort light pulses with an ensemble of quantum V-emitters

D. Ya. Bayramdurdyev¹, R. F. Malikov ^{✉1}

¹Bashkir State Pedagogical University Named After M. Akmulla,
3A Oktyabrskoy Revolyutsii Str., Ufa 450008, Russia

Authors

Davut Ya. Bayramdurdyev, e-mail: d.bayramdurdyev@gmail.com

Ramil F. Malikov, ORCID: [0000-0003-3457-0135](https://orcid.org/0000-0003-3457-0135), e-mail: rfmalikov@mail.ru

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Abstract. The paper reports the results of a theoretical study of the optical response of a two-dimensional supercrystal composed of quantum emitters with a doublet excited state (V-emitters) to a monochromatic external Gaussian pulse. The response dynamics are studied as a function of the input field's pulse area. It is shown that the field pulses interacting with the supercrystal lead to the generation of single, double, triple, and arc pulses with durations ten times shorter than those of the exciting pulses. The generation of single ultrashort pulses is shown to be independent of the magnitude of population excitation and can be realized in the presence of phase relaxation. The properties and effects of the optical response considered in this system suggest it may prove promising for applications in nanophotonics and quantum technologies.

Keywords: 2D superlattice, supercrystal, metasurface, three-level quantum emitters, quantum dots, self-oscillations, V-circuit, interaction of light pulses with 2D crystals, generation of optical pulses, phase relaxation

Introduction

Active research into metamaterials began following the discovery of graphene (Castro Neto et al. 2009; Novoselov et al. 2004). In recent decades, numerous reviews have been written on metamaterials and their applications in quantum photonics (Baimuratov et al. 2013; Bekenstein et al. 2020; Boneschanscher et al. 2014; Ding, Bozhevolnyi 2023; Solntsev et al. 2021; Soukoulis, Wegener 2010; Zheludev 2010). Ensembles of two- and three-level quantum systems represent the most widely used models for the theory of resonant laser-matter interactions. Within such models, foundational results have been obtained concerning various phenomena, such as photon echo (Kopvillem, Nagibarov 1963; Kurnit et al. 1964), self-induced transparency (McCall, Hahn 1967), electromagnetically induced transparency (Marangos 1998; Kasapi et al. 1995; Bloch 1946), optical nutation (Abragam 1963; Brewer, Shoemaker 1971), super-radiance (Bonifacio et al. 1971; Bonifacio, Lugiato 1975a; 1975b; Dicke 1954; Trifonov et al. 1979), and the coherent amplification of light pulses (Basov, Letokhov 1965; Frantz, Nodvik 1963; Hopf, Scully 1969; Kryukov, Letokhov 1970; Icsevgi, Lamb 1969; Varnavskii et al 1984), etc.

The discovery of new materials in the form of metasurfaces with lattices of regular quantum emitters (superlattices or supercrystals) has prompted intensive study of the optical response properties of supercrystals for two- and three-level electronic transition schemes (Basharov 1988; Vlasov et al 2013; Malikov, Malyshev 2017; 2025; Ryzhov et al 2019; 2021; Bayramdurdyev et al. 2020; Timoshchenko 2023).

In this paper, we investigate the nonlinear optical response of a supercritical structure consisting of ordered quantum emitters (quantum dots) with an excited-state doublet to an external field pulse. A pertinent example is semiconductor quantum dots with a degenerate valence band in a magnetic field (Efros et al. 1996). The high density and oscillator strength, along with the dipole-dipole interaction of the quantum emitters, play a significant role in the optical response of the supercrystal. Since the average dipole moment is directly dependent on the instantaneous quantum state, their interrelation is state-dependent. This relationship, combined with the intrinsic nonlinearity of the quantum emitter, provides positive feedback leading to highly stable dynamics, as observed in single layers of quantum emitters with ladder (Ryzhov et al. 2019) and lambda (Ryzhov et al. 2021) configurations. These systems exhibit monolayer responses characterized by self-oscillations, dynamic chaos, and high reflectivity within their operational frequency bands (Bayramdurdyev et al. 2020; Ryzhov et al. 2019; 2021).

Model

The energy level and transition scheme for isolated, identical V-type quantum emitters is shown in Fig. 1. It is assumed that a plane wave with a frequency quasi-resonant to the optical transitions of V-quantum emitters falls on the monolayer. Here, $|1\rangle$ is the ground state with energy ε_1 , $|2\rangle$ and $|3\rangle$ are the doublet states with energies ε_2 and ε_3 , respectively. Optically resolved transitions are $|1\rangle$ and $|2\rangle$, $|1\rangle$ and $|3\rangle$, characterized by transition dipole moments d_{21} and d_{31} . The excited states $|2\rangle$ and $|3\rangle$ spontaneously decay into the ground state $|1\rangle$ with decay constants γ_{21} and γ_{31} , respectively. Non-radiative relaxation within the doublet is accounted for by the constant γ_{32} .

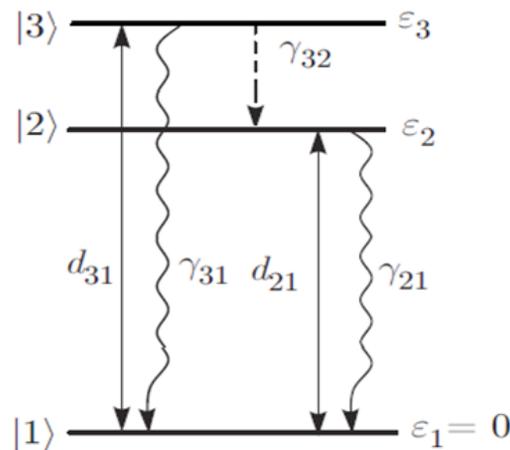


Fig. 1. Energy level diagram of a V-type quantum emitter

Basic equations

The optical dynamics of the quantum V-emitters within the supercrystal is described by the following system of equations for the density matrix (Bayramdurdyev et al. 2020):

$$\dot{\rho}_{33} = -(\rho_{31}\Omega^* + \rho_{31}^*\Omega) - (\gamma_{31} + \gamma_{32})\rho_{33}, \quad (1a)$$

$$\dot{\rho}_{22} = -\mu(\rho_{21}^*\Omega + \rho_{21}\Omega^*) - (\gamma_{21} + \gamma_{32})\rho_{33}, \quad (1b)$$

$$\dot{\rho}_{11} = \rho_{31}^*\Omega + \rho_{31}\Omega^* + \mu * (\rho_{21}^*\Omega + \rho_{21}\Omega^*) + \gamma_{31}\rho_{33} + \gamma_{21}\rho_{22}, \quad (1c)$$

$$\dot{\rho}_{32} = -i\Delta_{32}\rho_{32} - \Omega\rho_{21}^* - \mu\rho_{31}\Omega^* - 0.5(\gamma_{31} + \gamma_{21} + \gamma_{32})\rho_{32} - 2\Gamma \cdot \rho_{32}, \quad (1d)$$

$$\dot{\rho}_{31} = -i\Delta_{31}\rho_{31} + \Omega Z_{31} + \mu\Omega\rho_{32} - 0.5(\gamma_{31} + \gamma_{32})\rho_{31} - \Gamma \cdot \rho_{31}, \quad (1e)$$

$$\dot{\rho}_{21} = -i\Delta_{21}\rho_{21} + \mu\Omega Z_{21} + \Omega\rho_{32}^* - 0.5\gamma_{21}\rho_{21} - \Gamma \cdot \rho_{21}, \quad (1f)$$

where the dot above ρ_{ij} denotes the time derivative, Δ_{32} is the doublet splitting, $\Delta_{31} = \omega_0 - \omega_3$ and $\Delta_{32} = \omega_0 - \omega_2$ are the detuning frequencies of the external field ω_0 from the resonant frequencies of the transitions $3 \leftrightarrow 1$ and $3 \leftrightarrow 2$, respectively; $\mu = (\gamma_{21}/\gamma_{31})^{1/2}$, and Γ is the parameter corresponding to for the dephasing of the energy states. The diagonal elements of the density matrix ρ_{33} , ρ_{22} , ρ_{11} represent level populations, while the off-diagonal elements ρ_{31} and ρ_{21} correspond to the radiative transitions of the emitter, and ρ_{32} describes the coherence between the quantum radiative transitions. The radiation field is described by the Rabi amplitude Ω of the active field, which is the sum of the external field Ω_0 and the field radiated by all other quantum emitters at the location of a given emitter (the second term):

$$\Omega = \Omega_0 + (\gamma_R - i\Delta_L)(\rho_{31} + \mu\rho_{21}), \quad (2)$$

Here, γ_R and Δ_L are the amplitudes of the secondary field in the far and near zones, respectively. The first term determines the collective dynamic broadening, while the second determines the dynamic shifts of the quantum emitter energy levels, both of which depend on the population differences.

An optical oscillation generator

Figure 2 presents stationary solutions of equations (1)–(2). The left panel shows the dependence of the absolute Rabi amplitude $|\Omega|$ within the monolayer on the Rabi amplitude $|\Omega_0|$ of the external field. The right panel shows the dependence of the senior Lyapunov exponent of the solution on $|\Omega|$. In Figure 2 (b, c), for the case of monolayer excitation at the center of the V-quantum emitter’s doublet splitting ($\Delta_{31} = \Delta_{32}/2$, $\Delta_{32} = 40$, $\Delta_{21} = -20$), the dependence of $|\Omega|$ on $|\Omega_0|$ is unambiguous within a certain range of $|\Omega_0|$. However, for other considered values of the doublet splitting, the dependence of $|\Omega|$ on $|\Omega_0|$ is ambiguous, indicating multistability in the monolayer response of an isolated V-quantum emitter.

Let us consider the interaction of light pulses with the supercrystal when the quantum emitter system is in its ground state ($\rho_{33} = 0$, $\rho_{22} = 0$, $\rho_{11} = 1$). Excitation of the electronic system by an external field can be carried out on the second, third or at the center of the doublet splitting. In this study, we consider the case where the supercrystal’s electronic system is excited at the center of the doublet ($\Delta_{31} = \Delta_{32}/2$), corresponding to the stationary solution shown in Fig. 2 (b, c).

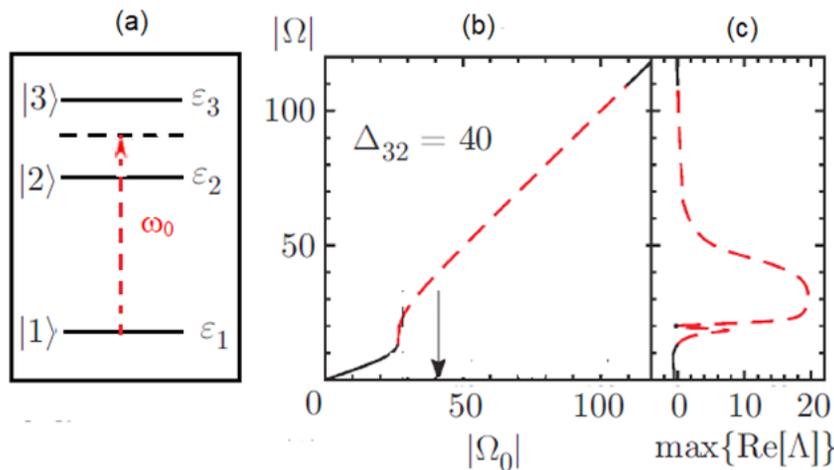


Fig. 2. Stationary solutions of equations (1)–(2)

We perform calculations for supercrystal parameters when the doublet width of the energy system is $\Delta_{32} = 40$. As follows from the stationary solutions, instabilities in the dynamic mode lead to the generation of oscillatory radiation, which eventually settles into the attractor (Fig. 3). The oscillatory optical response mode is realized at input field values in the range $|\Omega_0| = 26-100$.

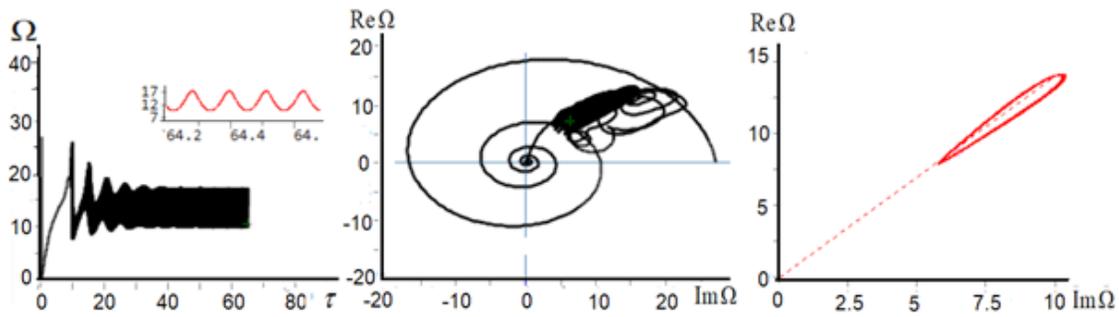


Fig. 3. Optical dynamics of a monolayer during the interaction of a field pulse with a supercrystal at the center of the doublet of V-quantum emitters ($\Delta_{31} = \Delta_{32}/2$). The field pulse magnitude is constant $Re\Omega_0 = 26, Im\Omega_0 = 0, g_{32} = 0.01, \Delta_{32} = 40, \Delta_{21} = -20, \Delta_L = 1000, \gamma_R = 100$

Figure 3 shows the dynamics of the field response and the corresponding phase portrait, illustrating an optical field generator. The system evolves into a periodic mode in the attractor (see inset); other modes of optical field generators can be found in (Malikov, Malyshev 2025). The first graph in Fig. 3 shows the response field dynamics $|\Omega(t)|$, the second shows the phase portrait of the dynamical system on the plane $(Re[\Omega], Im[\Omega])$, and the third shows the phase portrait of the attractor, which is a limit cycle. All quantities with dimensions of frequency are given in units of γ_{3p} , time is given in units of γ_{3l}^{-1}

Interaction of a short field pulse with a monolayer

Let us consider the interaction of short light pulses with a supercrystal when the quantum emitter system is in the ground state ($\rho_{33} = 0, \rho_{22} = 0, \rho_{11} = 1$), and the emitters are excited at the center of the doublet ($\Delta_{31} = \Delta_{32}/2, \Delta_{32} = 40, \Delta_{21} = -20$). The field pulse interacting with the supercrystal is defined as a Gaussian function with a temporal width τ_p .

Here, τ_0 is the time of the pulse maximum. The integral of this function is equal to the pulse area θ_0 , which in the coherent interactions is measured in units of π and is given by $\theta_0 = \int Re\Omega_0(\tau)d\Omega = \Omega_1\pi$. Figure 4 shows numerical solutions for different values of the pulse area interacting with the supercrystal, with a pulse width of $\tau_p = 1$.

In all subsequent figures, the first graph shows the input field pulse $|\Omega_0(t)|$ and the supercrystal response $|\Omega(t)|$; the second shows the dynamics of energy level populations; and the third shows the phase portrait on the plane $(Re[\Omega], Im[\Omega])$. All quantities with dimensions of frequency are given in units of γ_{3p} , time is given in units of γ_{3l}^{-1} .

As a result of numerical experiments, we found that when excited by a Gaussian pulse, the monolayer generates an optical response in the form of single, binary, ternary, etc. pulses. The optical response of the supercrystal in these modes becomes evident at pulse area values of $\theta_0 = 40\pi$. Moreover, the duration of the generated field pulses is ten times less than that of the exciting pulse. Studies with different doublet splitting values Δ_{32} have shown that stable generation of single, double, and triple pulses is observed even at lower values of the unput pulse area and duration.

The simulation results indicate that the duration of the generated pulses depends on the magnitude of the doublet splitting. Smaller splitting leads to shorter generated pulse durations. Within the duration of the exciting pulse, one, two, three, etc. pulses are generated, corresponding to periods of the optical generator. We cut out the corresponding oscillations from the optical pulse generator (Fig. 3) using an external field pulse of finite duration (3). As follows from the calculations, the optical response of the supercrystal takes the form of either a single pulse or a pulse cluster.

The generation of single pulses does not depend on the population state (ground or excited) of the overall system (see Fig. 5)

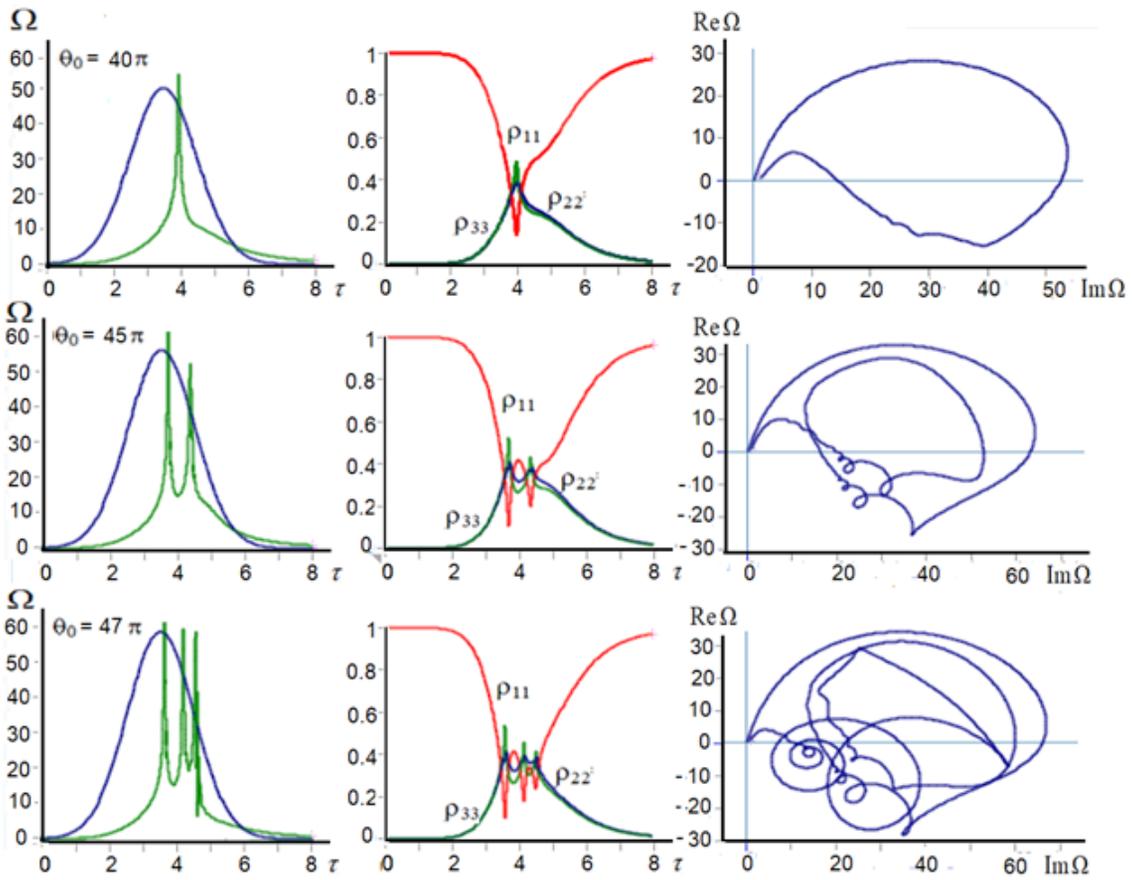


Fig. 4. Optical dynamics of a monolayer during the interaction of a field pulse of different areas with a supercrystal, with system excitation at the center of the V-quantum emitters ($\Delta_{31} = \Delta_{32}/2$), $\Delta_{32} = 40$, $\Delta_{21} = -20$ in the absence of phase relaxation $g_2 = 0$, $\tau_p = 1$, $\Delta_L = 1000$, $\gamma_R = 100$

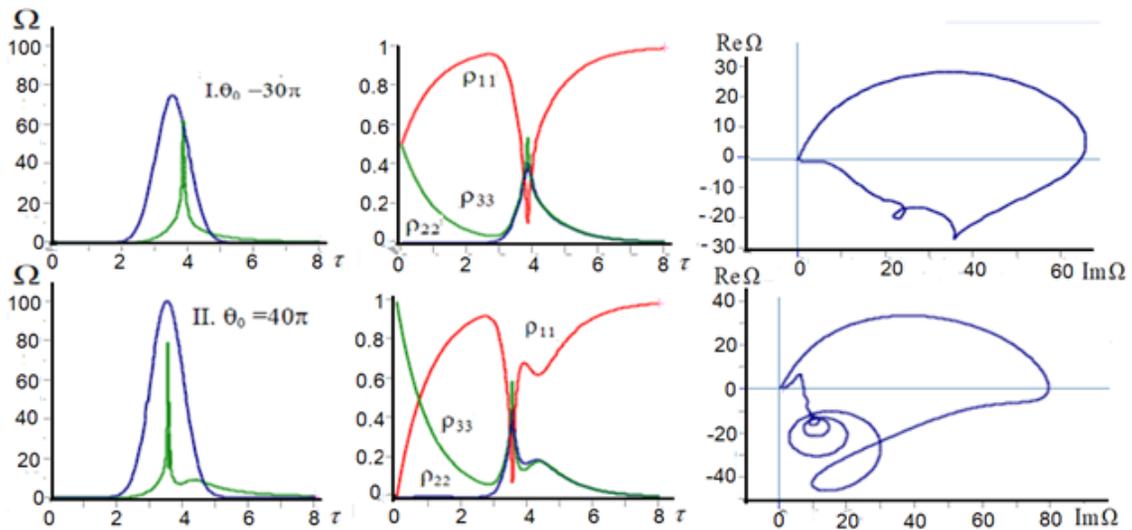


Fig. 5. Optical dynamics of a monolayer during the interaction of a field pulse with a supercrystal, with system excitation at the center of the doublet of V-emitters ($\Delta_{31} = \Delta_{32}/2$) in the absence of phase relaxation $g_2 = 0$, $\Delta_{32} = 40$, $\Delta_{21} = -20$. $\tau_p = 0.4$, $\Delta_L = 1000$, $\gamma_R = 100$ and different initial conditions
 I. $\rho_{33} = 0.5$, $\rho_{22} = 0.0$, $\rho_{11} = 0.5$. II. $\rho_{33} = 1.0$, $\rho_{22} = 0.0$, $\rho_{11} = 0.0$

The effect of phase relaxation on pulse generation

The influence of phase relaxation was studied for the three cases shown in Fig. 4. Figure 6 below presents numerical calculations of the effect of a short pulse on a supercrystal with phase relaxation $g_2 = 1$ and 1.2.

As the calculations show, phase relaxation affects the peak intensity and ultimately suppresses the single pulse generation.

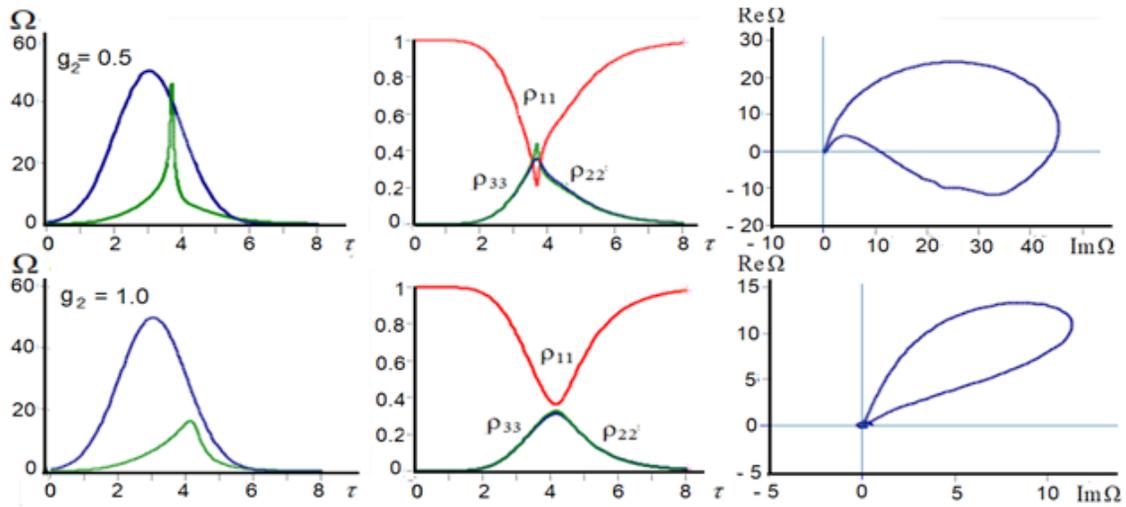


Fig. 6. Effect of phase relaxation on pulse generation by a supercrystal. The field pulse area is $\theta_0 = 40\pi$, with a duration of $\tau_p = 1$. The supercrystal parameters are $\Delta_{31} = \Delta_{32}/2$, $\Delta_{32} = 40$, $\Delta_{21} = -20$, $\Delta_L = 1000$, $\gamma_R = 100$

For the second case, where a binary pulse is observed, an increase in the magnitude of phase relaxation leads to the suppression and disappearance of the second pulse, i. e., the binary pulse is converted into a single pulse (see Fig. 7).

For the third case, shown in Fig. 4, an increase in phase relaxation initially leads to the formation of a pulse train, then to the fusion of these pulses, resulting in a shortened pulse with a significantly steeper leading edge compared to the incident Gaussian field pulse (Fig. 8).

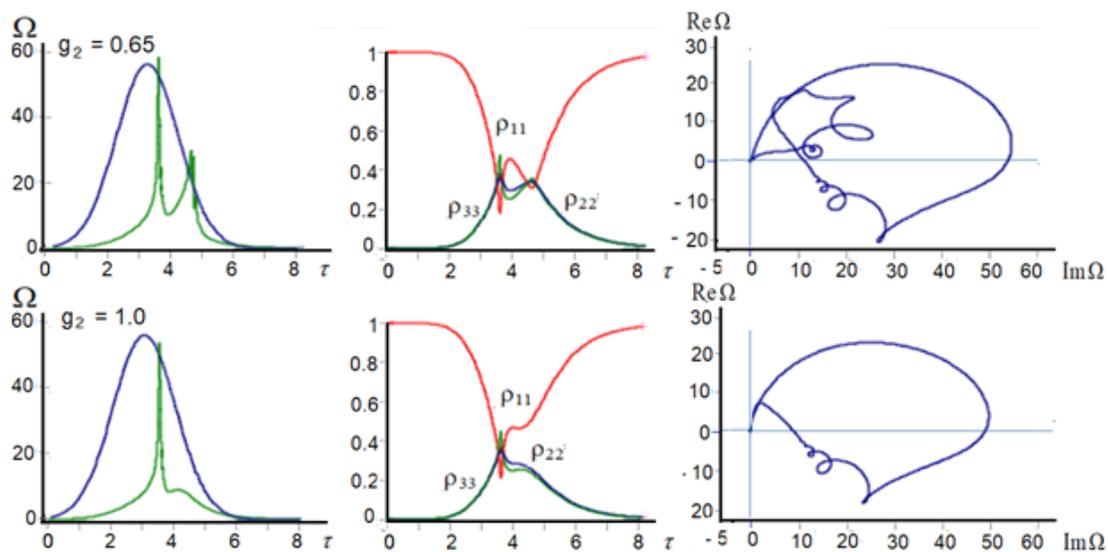


Fig. 7. Effect of phase relaxation on pulse generation by a supercrystal. The field pulse area is $\theta_0 = 50\pi$, with a duration of $\tau_p = 1$. The supercrystal parameters are $\Delta_{31} = \Delta_{32}/2$, $\Delta_{32} = 40$, $\Delta_{21} = -20$, $\Delta_L = 1000$, $\gamma_R = 100$

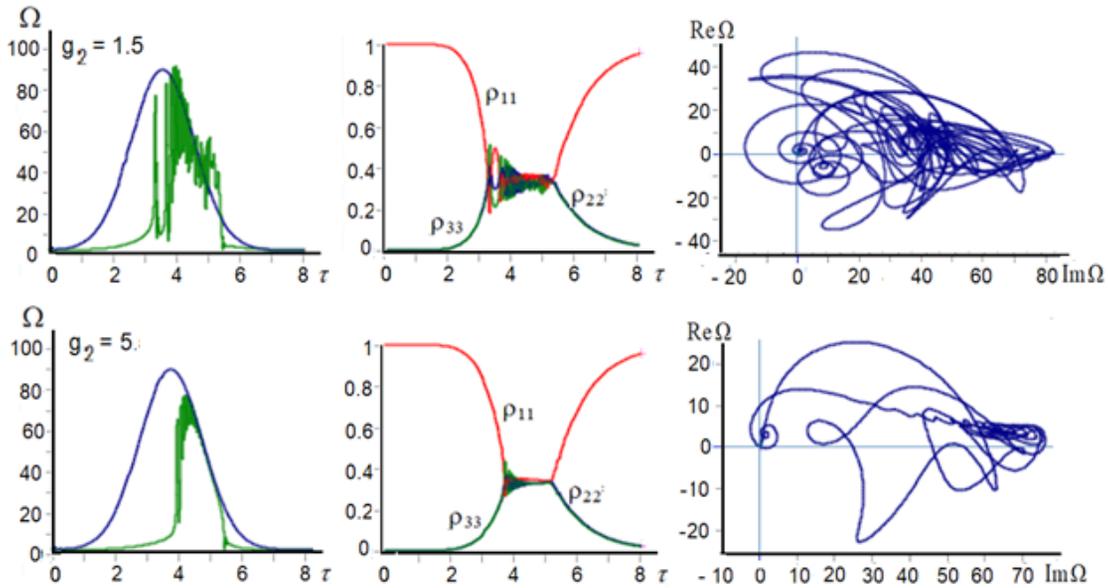


Fig. 8. Effect of phase relaxation on pulse generation by a supercrystal. The supercrystal parameters are $\Delta_{31} = \Delta_{32}/2$, $\Delta_{32} = 40$, $\Delta_{21} = -20$, $\Delta_L = 1000$, $\gamma_R = 100$

Conclusions

Previous studies (Basov, Letokhov 1965; Frantz, Nodvik 1963; Kryukov, Letokhov 1970; Içsevgi, Lamb 1969; Hope, Scully 1969; Varnavskii et al. 1984) examined the propagation of a light pulse through a dense absorbing (or amplifying) medium. In such a medium, the propagation of an ultrashort light pulse alters the entire population of the electronic subsystem, leading to various coherent effects — coherent absorption, coherent amplification, self-induced transparency, photon echo, and others. All of them depend critically on the sample length.

In contrast, the interaction of a light pulse with a constant field amplitude and a supercrystal results in the generation of an optical response in the form of self-oscillations and dynamic chaos (Bayramdurdyev et al., 2020).

When a Gaussian pulse interacts with a supercrystal, we observe the generation of single, double, and finite pulse trains whose durations are tens of times shorter than the exciting Gaussian pulse. This generation stems from the multistability and instability of the supercrystal’s electronic quantum subsystem (Bayramdurdyev et al., 2020), which arise from collective spontaneous emission (Arkhipov, Rozanov 2021), and, in our specific case, from both collective emission γ_R and the dipole-dipole interaction between quantum emitters Δ_L . Notably, the latter condition is an order of magnitude stronger than the collective interaction (superradiance) ($\Delta_L = 1000$, $\gamma_R = 100$).

The generation of a single ultrashort light pulse remains possible even in the presence of phase relaxation.

Thus, the results of this study on light pulse interaction with a supercrystal suggest that a V-type supercrystal can be recommended as a component in laser systems for generating single, binary, and train field pulses with durations tens of times shorter than of the input external pulse. The properties and effects of the supercrystal’s optical response discussed here indicate that an ordered system of V-type quantum emitters holds considerable promise for applications in nanophotonics and in optical-based quantum computing.

Conflict of Interest

The authors declare that there is no conflict of interest, either existing or potential.

Author Contributions

All authors made an equivalent contribution to the preparation of this publication.

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