

Ultrafast nonlinear spectroscopy with spatially confined fields

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Abstract. The combination of pulse shaping techniques for ultrashort laser pulses with nanoplasmonics leads to applications in the spatiotemporal control of electronic excitations with subwavelength precision. First, a method of finding shaped pulses for spatial selective excitation of individual nanostructures is presented. Second, double quantum coherence, a two-dimensional spectroscopic technique, yields the energies of single excitons and biexcitons and shows correlations between them. Through combining the spectroscopy with localization of optical fields we obtain more detailed information about the excitonic energies and couplings.

Keywords: Quantum dots, spatiotemporal control, pulse shaping, double quantum coherence, 2d spectroscopy

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INTRODUCTION

The techniques of controlling electronic excitations simultaneously in space (on a nanometer length scale) and time (on a femtosecond scale) become increasingly important in modern experiments [1, 2, 3]. Particularly, in spectroscopy methods progress in this field opens new possibilities for more specific knowledge about local properties of nanostructures [4, 5].

First we will present some spatially coupled nanostructures and pulse shaping setups for exciting one particular quantum system (quantum dot) out of three systems. In combination with the double quantum coherence (DQC) method [6] we will obtain more information than in conventional two-dimensional spectra.

SPATIOTEMPORAL CONTROL

Spatiotemporal control of electronic excitations with subwavelength precision and femtosecond time scale resolution can be achieved by combining pulse shaping techniques and nanoplasmonics [1, 2].

To find an appropriate geometrical layout we have to consider the main control mechanisms [2]: Plasmonic effects cause field enhancements in the near field depending on the structures. These effects interfere with each other and with the external field.

A typical example is the setup of a scanning tip over a sphere from [2] which uses different field enhancements for different polarization directions to control the excita-

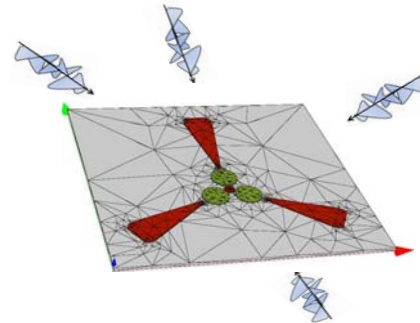


FIGURE 1. Geometry 1: Glass layer (bulk) and 2nm thick silver layers with varying triangle sizes and three 2nm thick InAs quantum dots with radius 20nm

tions between two positions in space. For our objectives we need to control the local excitation of more than two quantum dots which are close enough to couple and form new delocalized states.

We tested a plasmonic structure (Fig. 1) which consists of three silver triangles oriented in three directions to use polarization effects (based on [3]). The quantum dots are placed in close vicinity and between the sharp vertices and a central silver disc to bring the amplifications into the quantum dots. In order to address different triangles for different frequencies of the incoming pulse, the sizes of the triangles are varying.

To achieve full control of three points, one single incident pulse does not supply enough degrees of freedom.

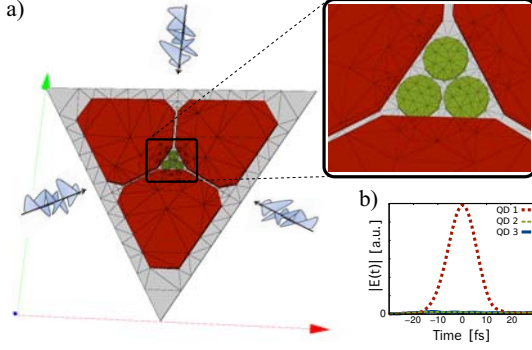


FIGURE 2. (a) Geometry 2: Glass layer (bulk) and 12 nm thick silver layers and three 4 nm thick InAs quantum dots with radius 20 nm, 120° rotationally symmetric, (b) time-dependent electric field distribution after optimization of the three quantum dots

We had to simulate the excitation with four independent pulse-shaped incoming pulses from different directions to obtain a sufficient control.

The structure for which we found the best spatiotemporal control is shown in Fig. 2(a). This waveguide-like structure keeps a full 120° symmetry where each of the three paths is illuminated by one independent shaped pulse. This setup is superior since there are no sharp vertices and the 120° symmetry helps to save computation time and avoids a position dependent chirp of the phase.

For the setup of Fig. 2(a), it is possible to generate a strong localization of the excitation at one quantum dot.

For the optimization of the field [2, 7] we use as input time harmonic solutions $\mathbf{E}_v(\omega, \mathbf{r})$ for each wavelength ω and for two perpendicular polarization directions $v = p, s$. They are calculated for one incoming direction using the Maxwell solver JCMsuite based on a time harmonic finite element method [8]. For shaping the pulse in time domain, we use a Fourier transform of these solutions

$$\mathbf{E}(t, \mathbf{r}) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} d\omega \sum_{v, \gamma} g_v^\gamma(\omega) \mathbf{E}_v^\gamma(\omega, \mathbf{r}) \quad (1)$$

and multiply with a weighting function

$$g_v^\gamma(\omega) = \sum_{n=1}^{20} f_v(\vartheta_n^\gamma) \frac{A_n^\gamma}{\sqrt{2\pi}} e^{-\frac{(\eta_n^\gamma - \omega)^2}{2(1/\sigma_n^\gamma)^2}} e^{i\omega\tau_n^\gamma} e^{i\phi_n^\gamma} \quad (2)$$

which represents a Fourier transformed composition of twenty Gaussian shaped pulses with amplitudes A , center times τ , inner frequencies η , widths σ , phases ϕ , and polarization angles ϑ for each pulse n , for each incoming direction γ and polarization v ($f_p = \cos$, $f_s = \sin$).

This method of composing electric fields reduces considerably the computational costs since all fields are calculated only once before the optimization.

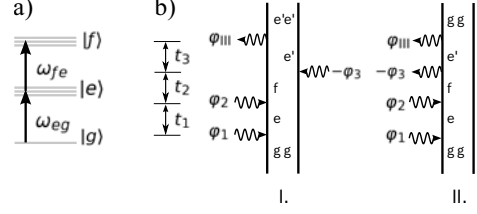


FIGURE 3. (a) Delocalized states with transition energy, (b) two possible pathways presented in a double-sided Feynman diagram

We shape a complex pulse form by superposing twenty different Gaussian shaped pulses with six variable parameters A , τ , η , σ , ϕ , and ϑ for each incoming direction γ and polarization direction v . These 360 parameters are optimized with the help of a genetic algorithm [7].

We start with three identical polarization shaped pulses and simulate the field distribution for the start pulse and 300 mutated copies. The pulse with the best value for the fitness function acts as parent for the new generation. The fitness function achieves a good quality for the pulse if the excitation of one quantum dot resembles a Gaussian-shaped target function in time domain and the other quantum dots have minimal excitation. The mean mutation step size is individually adaptive for all optimization parameters, depending on the improvement of the fitness function: higher improvement increases, lower improvement decreases the mutation step size. Within less than 3000 generations, we achieve a localization of the electric field at one quantum dot with field being a factor ten higher than at the other quantum dots (see Fig. 2(b)).

COUPLED EXCITONIC SYSTEM

Quantum dots placed close to each other exhibit coupled excitations, i.e. delocalized excitons due to Coulomb effects like Förster coupling even if we assume no wave function overlap [9, 10]. Through composing the local states $|i\rangle$ for a single exciton at quantum dot i and $|ij\rangle$ for the local state where quantum dots i and j are excited we obtain new delocalized states: a ground state g , single excitonic states e and biexcitonic states f (cf. Fig. 3(a)). These states can be obtained by diagonalizing the pure electronic part of the Hamiltonian [11].

COHERENT 2D SPECTROSCOPY

For coherent two-dimensional spectroscopy, time-ordered ultrashort pulses create single-exciton and double-exciton states in a well controlled way (cf.

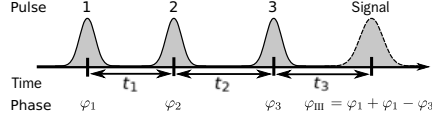


FIGURE 4. Pulse sequence for the DQC method

Fig. 4) [6]. In particular, for the DQC [5], all pulses of the sequence separated by delay time intervals are emitted in the same spatial direction. The signal with phase $\varphi_{\text{III}} = \varphi_1 + \varphi_2 - \varphi_3$ is detected. The different phases of the pulses allow us to separate different Liouville pathways in a phase-sensitive detector via the phase cycling technique [12]. The two possible pathways of excitation in our example are represented by the double-sided Feynman diagrams in Fig. 3(b) [13].

A 2d spectrum itself is obtained by Fourier transforming the third-order signal $S_{k_{\text{III}}} = S_{\text{I}} + S_{\text{II}}$ with respect to the times t_1 and t_2 , so that the frequency axes Ω_1 and Ω_2 represent the single and the double excitonic energy, respectively. The spectrum visualizes the energies of the electron in the ground, single excited, and double excited state and shows the coupling between them:

In Fig. 5(a) the absolute value of the full spectrum of three arbitrary coupled quantum dots with the corresponding energies of the single and biexcitons are shown.

Now, we combine this two-dimensional spectroscopy method with the possibility of high quality localization by only exciting one of the three quantum dots for the first pulse using the spatiotemporal control method introduced before.

In this case we obtain Fig. 5(b) providing new information: We find that each of the three peaks can be assigned to specific quantum dots. Here, the first pulse of the sequence only excites quantum dot 1. The localized spectrum shows almost only resonances for the transitions from e_2 to f_2 and from e_2 to f_3 . So we can say that only two of the three peaks arise from quantum dot 1. If we excite only quantum dot 2 instead, we find the resonance at the third peak of the full spectrum at e_1 to f_3 . Exciting only the remaining quantum dot 3 shows us that here the e_3 resonances play an important role (Figures not shown).

In conclusion, by combining nanoplasmonics and 2d coherent spectroscopy, we obtain new information such as spectral peaks connected to individual subsystems in a coupled nanostructure.

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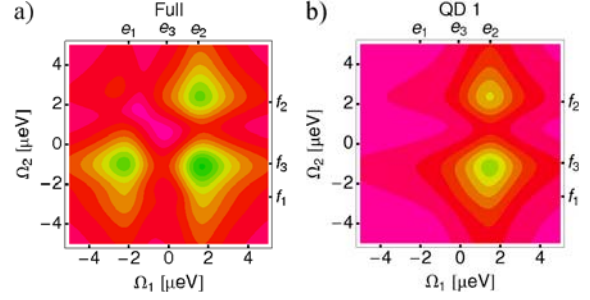


FIGURE 5. (a) Absolute value of the full k_3 spectrum, (b) absolute value of the localized k_3 spectrum (first pulse only excites one quantum dot) (both spectra given as detuning around the single (double) laser energy)

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