# Combining nanooptical fields and coherent spectroscopy on systems with delocalized excitons

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# ABSTRACT

For nanostructures such as semiconductor quantum dot emitters or biological systems like light harvesting complexes (photosynthesis) the coupling between individual constituents leads to the formation of delocalized exciton states. Coherent two dimensional spectroscopy is a versatile tool to investigate the structure of the excitonic states, whereas nanoplasmonics allows to localize optical fields on a nanoscale: We combine these two methods in a theoretical study and propose new experiments, such as the two dimensional spectra containing spatial resolution via localized fields. Using post processing of different spectra with localized fields, we can enhance certain spectroscopic features in standard coherent spectroscopy, e.g. by suppressing unwanted resonances.

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

# 1. INTRODUCTION

The spatial resolution of optical measurements with conventional light sources is limited by the wavelength of the incident light. Achieving electronic control below the diffraction limit opens a number of new methods in investigating nanosystems. Confining electronic excitations simultaneously on a nanometer length scale and on a femtosecond time scale becomes possible by combining nanoplasmonics with pulse shaping techniques.<sup>1-3</sup>

Nanoplasmonics uses different arrangements of metals or hybrids like nano antennas,<sup>4–6</sup> sharp triangles or metal tips<sup>2</sup> and exploits their frequency dependent properties. The metal nanostructures can be illuminated by polarization shaped short optical pulses that are formed with the help of a genetic algorithm<sup>7,8</sup> and simulated theoretically to obtain a detailed understanding of the local interaction.

On the other hand, multidimensional coherent spectroscopy yields a description of electronic excitations and couplings in complex molecular and nanoscale systems such as photosynthetic aggregates or semiconductor devices.<sup>9,10</sup> That provides also the basis for quantum computing, OLEDs or quantum effects in biological energy conversion.

The main idea of our work is to bring nanoplasmonics and coherent spectroscopy together to obtain new information on nanoscale materials. That means that we do coherent spectroscopy<sup>11</sup> with optimized shaped pulses to obtain more information than in ordinary two dimensional spectroscopy.

We begin with presenting the techniques of how to achieve spatiotemporal control in Sec. 2. Before we discuss the advantages and disadvantages of several tested geometries we introduce the underlying control mechanisms. We finish the first section by presenting some theoretical elements of our simulation methods. The following Sec. 3 gives a short introduction in two dimensional spectroscopy, especially in the double quantum coherence technique and explains the content of 2d spectra. That section also shows calculated signals. We conclude with the main idea in Sec. 4 by combining the methods presented in Sec. 2 and 3 to a "localized coherent spectroscopy".

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Figure 1: In this illustration of plasmonic structure for spatiotemporal studies silver particles are randomly distributed around four quantum dots. As explained in Sec. 2.2.1, this arrangement turns out to be inefficient.

## 2. SPATIOTEMPORAL CONTROL

A confinement of electronic excitations on length scales smaller than the wavelength of light is possible by combining nanoplasmonics with pulse shaping techniques<sup>1-3</sup> and will be discussed in more detail in the next section.

## 2.1 Control Mechanisms

In our setup we use combinations of different plasmonic materials with dielectrics to achieve field confinements. Resonant materials like silver, gallium arsenide or indium arsenide are simulated with the dielectric function of Ref. 12. Those arrangements produce enhanced electric field depending on the frequency of the incoming light.

Not only the optical material resonances, but also the form, size, and orientation of the nanostructures influence the spatial field distribution. All these effects interplay in the geometries described in Sec. 2.2.2, where triangles with different sizes and orientations are used: The sharp vertices of the tip are able to concentrate the field. With different sizes it is possible to address different optical frequencies. Finally, the position of the triangles plays an important role to use polarization effects of the polarization shaped pulses. Further discussion of such polarization effects can be found in Sec. 2.2.2.

A principal effect of confining excitation is the interference and enhancement of electromagnetic fields via local reflections and plasmon effects. Although all these fields interfere constructively or destructively we see that just one incoming pulse does not supply enough degrees of freedom. To achieve sufficient control of e.g. three coupled quantum dots we use three or more incoming pulses with individual shapes from different directions. Thus the fields of the incoming pulses can also interfere constructively or destructively at the sample. The specific simulation is discussed in Sec. 2.3.

## 2.2 Geometries

As stressed in the last section, the geometrical form of a setup strongly influences the field distribution. So, not all geometries are well-suited for an optimization process. In this section we will discuss how to find the optimized geometry.

There exist already some well-know setups, e.g. in Ref. 1–3, which are shown to control the excitation at two spatial positions in time. To investigate coupled quantum dots we like to control the excitation of three or more spatial points, which are in close vicinity. In particular, we focus on spatial points not be further apart than 50 nanometers.

## 2.2.1 Random geometries

In a naive approach one would start with a randomly arranged setup with intent to use as many of the discussed effects as possible. However, we found that this idea overestimates the possibilities of field control by shaping pulses via the used genetic algorithm and to design the pulses.

In randomly arranged geometries like that in Fig.1 we see that always some quantum dots are placed in a more preferential position, e.g. they are closer at metal structures with high field amplifications. It turns out that localization of the electric field has a bad quality for quantum dots at disadvantaged positions (e.g. if the quantum dot is not close enough to a metal structure). To illustrate the principle of localized spectroscopy, we will concentrate on symmetric arrangements in the following.



Figure 2: (a) A setup of three silver triangles and a silver cylinder on a glass layer (without quantum dots) serves to present polarization effects. The polarization direction of the incoming plane wave can be regulated. (b) The field distribution of the layout depends on the incoming polarization direction. On the left we use s-polarized, on the right p-polarized light.

#### 2.2.2 Antenna-like geometries

This kind of geometry affords a good study of polarization shaping effects because of the different orientations of the antennas. The first simple setup is shown in Fig. 2a. We use again only one polarization shaped incoming pulse that incides perpendicular from above. In general we can say that the largest field enhancements occur always on those boundaries of metal structures, which lie in the direction of the polarization vector. This effect is transferred to the triangles given in Fig. 2b. On the right of this picture we see the field distributions for two perpendicular polarization directions: on the left hand side the s- and on the right hand side the p-polarization. Placing the quantum dots between the tip and the center circle, we can excite two of three quantum dots simultaneously. However, the calculations show, that the ratio between the excitation strength of the two quantum dots and the excitation of the weak excited quantum dot is not high enough for our purpose.

We tried to reach a better ratio by implementing each triangle with different size to have different resonance frequencies. We also deformed the triangles, rounded the vertices or replaced them by rods to change the optical properties of the nanoparticles.<sup>5,13</sup> So far these ideas provide some enhancement, but during our attempts, we did not achieve sufficient localization for this structures. Displacing them to an antisymmetric setup like in Fig. 3a led to the same problems as discussed in Sec. 2.2.1: only for spatially advantaged dots a localization is possible.

#### 2.2.3 Waveguide-like geometries

The layout that satisfies our requirements best is shown in Fig. 3b. A metal structure similar to a waveguide inspired by nanoantennas, that can guide electric field strength into the nanostructures. This setup is discussed in the following section.

## 2.3 Simulations

For a theoretical simulation of the field localization we use the time harmonic results from the Maxwell solver of JCMsuite.<sup>14</sup> Maxwell's equations are solved in frequency domain using finite elements.

We evaluate the electric fields  $E_{\nu}^{\gamma}(\omega, \mathbf{r})$  at the center of the quantum dots for all incoming directions  $\gamma$  and the polarization directions  $\nu = p, s$ . For controlling the field distributions in time domain, we apply a Fourier transformation:

$$\boldsymbol{E}(t,\boldsymbol{r}) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} d\omega \,\mathrm{e}^{\mathrm{i}\omega t} \,g_{\nu}^{\gamma}(\omega) \,\boldsymbol{E}_{\nu}^{\gamma}(\omega,\boldsymbol{r}), \tag{1}$$

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Figure 3: (a) The geometry of an 3d asymmetric ray-like layout with InAs quantum dots and silver rods and disks (top view) shows different optimization qualities depending on which quantum dot is excited. (b) This three-armed waveguide-like setup satisfies our requirements. The three InAs quantum dots on a GaAs layer have a radius of 10 nm and a distance of about 40 nm and they are surrounded by three silver polygons with 250 nm for the longest edge. They were illuminated by three individual polarization shaped pulses with incoming directions parallel to the waveguide arms. The field strength is measured in the center of the quantum dots (white crosses). (c) The absolute value of the electric field strength in time domain is measured at the three quantum dots in Fig. 3b. It shows the optimized distribution for exciting only quantum dot 1.

where g is a weighting function, that enables us to shape the incoming pulses. The function g represents a Fourier transformed composition of n Gaussian shaped pulses (similar to Ref. 7):

$$g_{\nu}^{\gamma}(\omega) = \sum_{n} f_{\nu}(\vartheta_{n}^{\gamma}) \frac{A_{n}^{\gamma}}{\sqrt{2\pi}} e^{-(\eta_{n}^{\gamma}-\omega)^{2} \sigma_{n}^{\gamma^{2}/2}} e^{\mathrm{i}\omega\tau_{n}^{\gamma}} e^{\mathrm{i}\varphi_{n}^{\gamma}}, \qquad (2)$$

with  $f_p = \cos$  and  $f_s = \sin$ . Thus it is possible to simulate the incoming electric field with a fixed amplitude and to weight each frequency individually only by changing the parameters of g: the polarization angle  $\vartheta$ , the amplitudes  $A_n^{\gamma}$ , the center times  $\tau_n^{\gamma}$ , inner frequencies  $\eta_n^{\gamma}$ , widths  $\sigma_n^{\gamma}$  and the phases  $\varphi_n^{\gamma}$ . A number of n = 20pulses is enough to form the pulses sufficiently.

Composing the electric fields in this way reduces considerably the computational costs since all fields have to be simulated only once. In contrast to a time domain solver, in frequency domain there is no new simulation needed for altered incoming pulses.

In order to obtain the shaped pulses that satisfy our requested field distribution we use a genetic algorithm<sup>1, 7, 8</sup> that optimizes the six parameters of the weighting function g. Three incoming directions and twenty composed Gaussian pulses results in  $6 \cdot 3 \cdot 20 = 360$  parameters to be optimized simultaneously. The algorithm is explained in more detail in the appendix.

In less than 10000 generations it is possible to see if the chosen geometry provides a highly localized excitation. In the geometry described in Sec. 2.2.3 we achieve the localization of the electronic excitation in time domain shown in Fig. 3c.

# 3. 2D SPECTROSCOPY

We will present a special kind of coherent spectroscopy, the double quantum coherence<sup>9</sup> (DQC). This third-order nonlinear optical experiment uses a pulse sequence that is shown in Fig. 4a. Three time-ordered pulses with variable time delays  $t_1$ ,  $t_2$ , and  $t_3$  create a signal in the sample. This signal is mixed with a fourth pulse, the so called local oscillator. This is called heterodyne detection.<sup>9</sup>

Since the dimensions of the structures which are small compared to the wavelength of the pulses used, all the incoming pulses come from the same spatial direction. This disadvantage in signal selectivity can be compensated



Figure 4: (a) All pulses have the same direction but different phases to use the phase cycling technique. Only the phase  $\varphi_1 + \varphi_2 - \varphi_3$  is detected. (b) The two-sided Feynman diagram shows the possible Liouville pathways of the DQC method.

by different phases for the pulses.<sup>11</sup> To separate the pulses, we make use of the phase cycling technique.<sup>15</sup> Here, the experiment is repeated several times with varied phases  $\varphi_1$ ,  $\varphi_2$ , and  $\varphi_3$ , while a phase  $\varphi_{\text{III}} = \varphi_1 + \varphi_2 - \varphi_3$  is extracted from the signal.

The first pulse of the sequence creates a single exciton (single quantum coherence), the second creates a double excitonic state (double quantum coherence) and the third one again a single excitonic state. The possible pathways of excitation are represented by double-sided Feynman diagrams (cf. Fig. 4b).<sup>16</sup>

In a two dimensional spectrum not only energies of single and biexcitons but also the correlations between their states become visible. A 2d spectrum is obtained by Fourier transforming the signal  $S(t_1, t_2, t_3)$  with respect to the time differences  $t_1$  and  $t_2$  at a fixed time difference  $t_3$ . Thus the signal  $S(\Omega_1, \Omega_2)$  now depends on the single and biexcitonic frequencies  $\Omega_1$  and  $\Omega_2$ , respectively. The standard DQC signal  $S_{k_{\text{III}}} = S_i + S_{ii}$  for a heterodyne detection reads:<sup>9</sup>

$$S_{i}(\Omega_{1},\Omega_{2},t_{3}) = \frac{1}{\hbar^{3}} \sum_{ee'f} \mu_{e'f} E_{4}^{*}(\omega_{fe'}) \mu_{ge'} E_{3}^{*}(\omega_{e'g}) \mu_{ef}^{*} E_{2}(\omega_{fe}) \mu_{ge}^{*} E_{1}(\omega_{eg}) \frac{e^{-i\xi_{fe'}t_{3}}}{(\Omega_{2} - \xi_{fg})(\Omega_{1} - \xi_{eg})}$$
(3)

and

$$S_{\rm ii}(\Omega_1, \Omega_2, t_3) = -\frac{1}{\hbar^3} \sum_{e'fj} \mu_{ge'} E_4^*(\omega_{e'g}) \mu_{e'f} E_3^*(\omega_{fe'}) \mu_{ef}^* E_2(\omega_{fe}) \mu_{ge}^* E_1(\omega_{eg}) \frac{\mathrm{e}^{-\mathrm{i}\xi_{e'g}t_3}}{(\Omega_2 - \xi_{fg})(\Omega_1 - \xi_{eg})},\tag{4}$$

with the Fourier transformation of the electric field envelopes  $E_i(\omega)$  for the pulses i = 1, 2, 3, 4, the dipole matrix element  $\mu_{ij}$  for the transitions from exciton state j to i, the frequencies  $\omega_{ij} = \omega_i - \omega_j$ ,  $\omega_i$  the exciton energy, and  $\xi_{ij} = \omega_{ij} + i\gamma_{ij}$  with the damping  $\gamma_{ij}$ . A corresponding spectrum is shown in Fig. 5b. The frequencies are detuned around the single and double gap frequency, respectively.

#### 4. LOCALIZED SPECTROSCOPY

We demonstrate the localized spectroscopy method for three two-level quantum dots. A local basis consists of the states  $|i\rangle_1$  for each quantum dot *i*. Placing the dots close to each other yields new states. These delocalized states are caused by electronic coupling between quantum dots due to Coulomb effects like Förster coupling.<sup>17–19</sup> Note, that coupled excitations are possible even if we assume that the wave function do not overlap. By diagonalizing the pure electronic part of the Hamiltonian we get the new states:<sup>20,21</sup> a ground state *g*, three single excitonic states *f* (shown in Fig. 5a). The delocalized basis, consisting of the states *g*, *e*, and *f*, can be written as a composition of the local basis.

In Fig. 5b we show the imaginary part of a calculated spectrum of three coupled quantum dots. For the eigenenergies and the coupling strengths typical values for self-organized Ga/InAs quantum dots<sup>17</sup> are chosen



Figure 5: (a) Three single two-level quantum dots form delocalized states if their distance is small enough. (b) Two-dimensional spectrum of the coupled quantum dots are shown here: the complete absolute value of the DQC signal, (c) the same spectrum for the case that the first pulse of the sequence excites perfect just one quantum dot. The frequencies are given as detuning relative to the single and double gap frequency.

(for geometries presented in Fig. 3b). For a given resonance, we can read the single exciton state  $e_1$ ,  $e_2$ , and  $e_3$  on the  $\Omega_1$ -axis and the two exciton energy for states  $f_1$ ,  $f_2$ , and  $f_3$  on the  $\Omega_2$ -axis. Each resonance shows the correlation between the single and two exciton states. The strongest resonances occur at  $e_2$  with  $f_2$  and at  $e_2$  with  $f_3$ . Also a strong coupling between  $e_1$  and  $f_3$  is seen.

While in Fig. 5b all pulses of the sequence excite all three quantum dots simultaneously, in Fig. 5c the first pulse of the sequence only excites quantum dot 1. This field design is possible with help of the techniques discussed in the previous sections. Here, we choose the first pulse to be localized since the first pulse creates a single excitonic state after starting from ground state in the Liouville pathway (cf. Fig. 4b).

We see that the localized spectrum is different from that without localization. The strong resonances for the transitions from  $e_2$  to  $f_2$  and to  $f_3$  also appear, but there is no peak at  $e_1$  to  $f_3$ . The energy axis  $\Omega_1$  is connected to the energy difference of the ground to single exciton coherence after the first pulse. So we conclude that the third peak does not arise from a transition at quantum dot one during the first pulse. Which quantum dot is responsible for the  $e_1$  to  $f_1$  peak becomes visible by looking at the other localized spectra (not shown here). We find that this resonance arises from pathways localized at step 1 at quantum dot 3.

Obviously localized, coherent spectra provide new information. With their help we are able to connect each resonance to a specific quantum dot. We will use this information for more detailed spectroscopy of nanoscale coupling between individual emitters in the future.

# APPENDIX A. GENETIC ALGORITHM

A set of *n* optimization parameters can be expressed as a *n*-dimensional vector *a*. In every generation 300 mutated copies  $a_{\text{child}}$  of the parent pulse  $a_{\text{parent}}$  are created by adding the parameter dependent mutation step size  $\Delta a$  multiplied with a Gaussian distributed random number  $\chi$  for every of the 360 variation parameters *a*:

$$a_{\rm child} = a_{\rm parent} + \chi \cdot \Delta a \tag{5}$$

Next, the best child is selected. This is done by evaluating a scalar cost function for every child and selecting the one with the smallest cost value. We choose a cost function that compares the electric field distribution in the center of the excited quantum dot caused by the pulse with a target distribution.

We have one condition for each quantum dot. While the electric field distribution at position  $r_1$  should match with a Gaussian shaped excitation, the field distribution at positions  $r_2$  and  $r_3$  should be minimal.

The requested time dependence of the absolute value of this electric field distribution at the excited dot at  $r_1$  is given by a target function

$$\tan(t) = B e^{-(t-T)^2/\epsilon^2}$$
 (6)

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that exhibits a Gaussian shaped excitation with an amplitude B that depends on the chosen geometry, a time T at which the excitation is centered, and an excitation width  $\epsilon$ . Since the other quantum dots should not be excited the target function for positions  $r_2$  and  $r_3$  is identically zero.

To account larger deviations stronger than smaller ones we calculate the square of the absolute value of the difference between the target and the actual distribution. Afterwards we integrate over all times so that a higher integration result means a overall worse matching.

We tested different ways of combining all three conditions to obtain one value that represents the quality of the field distributions at all positions. It turned out that all conditions are fulfilled most uniform if we minimize a cost function given by:

$$\operatorname{cost}(\operatorname{child}) = \max\left(\int_{-\infty}^{\infty} |E_{\operatorname{child}}(t, r_1) - \operatorname{tar}(t)|^2 dt, \int_{-\infty}^{\infty} |E_{\operatorname{child}}(t, r_2)|^2 dt, \int_{-\infty}^{\infty} |E_{\operatorname{child}}(t, r_3)|^2 dt\right) \stackrel{!}{=} \min.$$
(7)

The child with the lowest value of the cost function acts as parent pulse for the next generation.

We use an adaptive mutation step size  $\Delta a$  that varies depending on the average of the step sizes of the previous 100 children.<sup>8</sup> Thus we quickly approach the minimum of the cost function and reach the lowest values with finer steps.

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