Reconstruction of the wave functions of coupled nanoscopic emitters using a coherent optical technique

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We show how coherent, spatially resolved spectroscopy can disentangle complex hybrid wave functions into wave functions of the individual emitters. This way, detailed information on the coupling of the individual emitters, not available in far-field spectroscopy, can be revealed. Here we propose a quantum state tomography protocol that relies on the ability to selectively excite each emitter individually by spatially localized pulses. Simulations of coupled semiconductor GaAs/InAs quantum dots using light fields available in current nanoplasmonics show that undesired resonances can be removed from measured spectra. The method can be applied to a broad range of coupled emitters to study the internal coupling, including pigments in photosynthesis and artificial light harvesting.

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I. INTRODUCTION

The formation of collective optical resonances from Coulomb-coupled optical emitters is a very general phenomenon, including examples from chromophores in biological light harvesting complexes,1–4 semiconductor quantum dots,5,6 metal nanoparticles, and composite systems, such as plasmon lasers.7

For all these structures, dipole-dipole coupling occurs on a nanometer scale and the states of the individual emitters hybridize to form new collective, so-called excitonic states, delocalized over the whole structure. Far-field excitation, governed by the wavelength resolution limit \( \lambda/2 \), can probe only delocalized exciton states of a nanostructure. Related far-field experiments such as absorption, pump probe, and four-wave mixing are unable to disentangle the individual contributions of the coupled emitters from the collective optical response, because the exciting fields are spatially constant on the scale of the entire structure and cannot discriminate different emitters. In contrast, spatially local spectroscopy such as near-field spectroscopy can, in principle, address the individual emitters.

In this paper, we propose a new class of measurements that combine coherent nonlinear spectroscopy with near-field optics to reconstruct the contributions of single emitters to the delocalized wave function in a spatially extended nanostructure. As an example, we demonstrate how a coherent double-quantum-coherence optical technique8 may be combined with spatially localized fields to reconstruct the exciton wave functions of three dipole coupled self-organized GaAs/InAs quantum dots. This constitutes a particular quantum state tomography. The presented procedure is independent of the technique for localizing the fields at individual emitters. Several localization methods are known and are already applied to a broad range of nanoemitting structures, for example, using (metallized) near-field fiber tips,5,8,10 metal tips,11,12 nano antennas,13–17 and metal structures combined with pulse-shaped fields.18,19

Quantum state tomography is a development aimed at the direct reconstruction of wave functions or more generally the density matrix, first proposed by Fano.20 The importance of quantum state tomography results from the fact that the reconstruction and knowledge of the wave function opens the possibility of calculating new observables not related to optics at all. Examples include magnetic moments and transport properties. So far, wave functions are seldom directly accessible by experiments.21 Recent advances include imaging of single orbitals using soft x-ray pulses22,23 and the reconstruction of states.24,25 Applications so far include spin-1/2 particles,26 photon states using the Wigner function,27,28 vibrational states,29 and Josephson junctions.30 In contrast to earlier approaches, the quantum state tomography developed in this paper combines optical fields, highly localized in time and space with coherent two-dimensional (2D) spectroscopy, using a sequence of light pulses with controlled envelopes and phases.31,32

II. EXCITONS IN COUPLED NANOSTRUCTURES

As a typical example for coupled nanostructures with delocalized wave functions, we study three coupled self-organized semiconductor quantum dots5,33,34 [Fig. 1(a)]. The quantum-dot distance is assumed to be sufficiently large to have no electronic wave function overlap between the quantum dots. In this case we study interdot coupling in the form of dipole-dipole (or Förster) coupling known from self-organized GaAs/InAs quantum dots: Parameters like dot size, dot distances, coupling constants, and energy shifts are well known from theory35 and experiments.36 Each quantum dot is represented as a two-level system. This is a valid assumption for quantum dots provided quantum dots (i) have no spin-orbit splitting and a sufficiently large biexcitonic shift, (ii) are negatively charged, or (iii) have spin-orbit coupling bigger than the inter-quantum-dot couplings.37,38 For self-organized quantum dots with sizes of 20 nm and...
interdot distances around 40 nm, the dipole coupling is about several μeV with a Lorentzian zero phonon line (ZPL) width of γ = 1 μeV at low temperatures (e.g., T = 4 K).\textsuperscript{38,39} We neglected the influence of the phonon side bands, since their amplitude in the spectra is one to two orders smaller than the amplitude of the ZPL resonance at low temperatures.\textsuperscript{38,39}

Three coupled quantum dots exhibit joint states: a ground state \( g \); three single-exciton states \( e_1, e_2, \) and \( e_3 \); and three two-exciton states \( f_1, f_2, \) and \( f_3 \) [Fig. 1(b)]. The system has one triexciton state, but these states are of no relevance here.\textsuperscript{38,39}

The ground state of the uncoupled quantum dots is not changed by the induced dipole-dipole coupling. The delocalized single-exciton states \( |e⟩ \) resulting from the dipole-dipole interaction are composed of local, uncoupled quantum-dot states \( |i⟩ \) (quantum dot \( i \) in excited state): \( |e⟩ = \sum c^i_e |i⟩ \). \( |e⟩ \) is an energy eigenstate of the coupled quantum-dot system, \( c^i_e \) the expansion coefficients. Similarly, two-exciton states \( |f⟩ \) are composed of states with two local excitations at quantum dots \( i \) and \( j \): \( |f⟩ = \sum_{i,j} c^i_j f^i_j |i,j⟩ \). In general, excited states of \( N \) coupled two-level system emitters form a ground state \( g \), \( N \) delocalized single-exciton states \( e \), and \( N(N − 1)/2 \) delocalized two excitons states \( f \). For our three-dot case, we choose couplings between two quantum dots slightly stronger than to the third quantum dot (parameters given in Table I). Here, \( H_0 \) includes along the diagonal the transition-frequency local emitters modified by single- and two-exciton shifts, respectively. The off-diagonal elements describe interactions of excitation transfer caused by, for example, dipole-dipole interactions.

![FIG. 1. (Color online) (a) Three dipole-dipole coupled self-organized InAs quantum dots. (b) Exciton level scheme of the three coupled quantum dots.](image)

**TABLE I.** Hamilton operator in matrix form. (a) The single-exciton block and (b) the two-exciton block. All values are given in μeV. The diagonal elements of the matrices are given as detuning to a mean gap frequency, (a) \( \omega_{\text{gpp}} \) for the single excitons and (b) \( 2\omega_{\text{gpp}} \) for the two excitons, with \( \omega_{\text{gpp}} = 1.053 \) eV and the two-exciton shifts \( V_{11} = 0.1 \) μeV, \( V_{12} = -2.5 \) μeV, and \( V_{13} = -1.5 \) μeV.

| \( |i⟩|H_0⟩|j⟩ \) | 1 | 2 | 3 |
|---|---|---|---|
| (a) 1 | 2.0 | 1.0 | 0.2 |
| 2 | 1.0 | 0.2 | 0.1 |
| 3 | 0.2 | 0.1 | -2.5 |
| (b) \( |i⟩|H_0⟩|k⟩ \) | 1.2 | 1.3 | 2.3 |
| 1,2 | \( |1⟩|H_0⟩|1⟩ + |2⟩|H_0⟩|2⟩ + V_{11} \) | \( ⟨3⟩|H_0⟩|2⟩ \) | \( ⟨3⟩|H_0⟩|1⟩ \) |
| 1,3 | \( 2|H_0⟩|3⟩ \) | \( ⟨1⟩|H_0⟩|1⟩ + ⟨3⟩|H_0⟩|3⟩ + V_{21} \) | \( ⟨2⟩|H_0⟩|1⟩ \) |
| 2,3 | \( |1⟩|H_0⟩|3⟩ \) | \( |1⟩|H_0⟩|2⟩ \) | \( ⟨3⟩|H_0⟩|1⟩ + ⟨3⟩|H_0⟩|3⟩ + V_{31} \) |

![FIG. 2. Absorption spectrum coupled (solid) and uncoupled (dashed) quantum dots. The detection frequency \( \omega \) is given as detuning relative to frequency \( \omega_{\text{gap}} = 1.053 \) eV (transition frequency of uncoupled quantum dot 3).](image)

First, to characterize the system within far-field spectroscopy, we calculate the linear absorption spectrum:

\[
\alpha(\omega) \propto \sum_e \frac{|\mu_{ee}|^2}{(\omega - \omega_{ee})^2 + \gamma^2}.
\]

Here, \( \mu_{ee} \) is the dipole moment for ground-state to single-exciton transition, \( \omega_{ee} \) is the transition frequency, and \( \gamma \) is the dephasing constant.

The absorption spectrum of the coupled quantum-dot structure is plotted in Fig. 2 (solid). The single-exciton states \( e_1, e_2, \) and \( e_3 \) overlap spectrally such that only \( e_1 \) and \( e_2 \) are well resolved and \( e_3 \) contributes only with a spectral shoulder. By comparing coupled and uncoupled (dashed) spectra, one recognizes that the oscillator strength is originally evenly distributed but strongly modified, since the dipole-dipole coupling forms excitons delocalized over the entire structure.

**III. INGREDIENTS FOR RECONSTRUCTING DELOCALIZED STATES**

Our main goal is to gain information on the buildup of the delocalized wave functions of the excitonic states (i.e., on the expansion coefficients \( c^i_e \)) for a given single-exciton state \( |e⟩ \).
For this purpose, we use coherent, spatially local spectroscopy composed of three ingredients:
(i) local nanoscale excitation provided by metallic nanoantennas and refined pulse-shaping techniques to optically address individual quantum dots (Sec. III A),
(ii) phase cycling of the optical response to disentangle the total nonlinear response into desired quantum paths (Sec. III B), and
(iii) a postprocessing procedure to calculate the coefficients \( c_i^j \) (Sec. IV).

In general, (ii) and (iii) can be applied to any quantum system representable by spatially separated coupled emitters, if any localization technique (i) is available.

### A. Localized excitation

A main ingredient of our scheme is the local excitation of individual quantum dots. In our specific example, we achieve local excitation of the individual quantum dots by a plasmonic antenna structure of triangular symmetry on a subwavelength scale [Fig. 3(a)]. These metal structures can be realized by e-beam lithography. Solving Maxwell’s equations for this geometry shows that plasmonic effects and an optimization procedure of the applied pulses allows us to selectively excite single quantum dots.

For optimizing the pulse envelope of a single pulse \( E(t, r) \) towards a field localization at only one quantum dot, we use time-harmonic solutions \( E_v(\omega, r) \), represented by incident plane waves of polarization directions (superscripts \( p \) and \( s \)) and incoming direction (indexed as \( v \)).

\[
E(t, r) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} d\omega \sum_v g_v(\omega) E_v(\omega, r) e^{-i\omega t}. \tag{2}
\]

Pulse shaping is introduced by the weighting function:

\[
g_v(\omega) = \sum_n f^v(\theta_n) \frac{A_n}{\sqrt{2\pi}} e^{-(\eta_v-\omega)^2/2+\text{cis}\sigma_n+i\beta}, \tag{3}
\]

which represents a composition of Gaussian pulses with amplitudes \( A \), center times \( \tau \), frequencies \( \eta \), widths \( \sigma \), phases \( \beta \), and polarization angle \( \theta \) for each pulse \( n \) projected to polarization direction \( v \) (\( f^p = \cos \), \( f^s = \sin \)). \( g_v(\omega) \) has to be determined by optimization. To increase the number of optimization parameters, we combine the three incoming pulses from three directions, using 120° symmetry of the sample. For this paper, details of the optimization procedure are of no relevance but can be found in Refs. 40 and 44. Later on, the absolute value of \( E(t) \) in the quantum-dot centers is the input for the calculation of the localized spectra.

In Fig. 3(b), the spatial field distribution for the optimized total field around the quantum-dot transition frequency is shown. It can be recognized that a chosen, single quantum dot is excited stronger than the other quantum dots. We observe field enhancements between different quantum dot sites of a factor of eight or larger. Note that the optimized fields in frequency domain show that polarization and propagation phase effects cause localization and not a frequency-based selection of different quantum dots.

Note that the presented localization scheme using excitation pads is just an example. For application of the protocol to other systems, other spatial localization schemes might be used.

### B. Phase cycling detection of coherent signals

As explained in Sec. III A, a sequence of three spatially optimized pulse envelopes \( E^l \) with phases \( \varphi_l \) and laser frequency \( \omega_l \) is used,

\[
E(r, t) = E^1(t, t - t_1 - t_2 - t_3) e^{i(\omega_1(t-t_1-t_2-t_3)+\varphi_1)} + E^2(t, t - t_1 - t_2 - t_3) e^{i(\omega_2(t-t_2-t_3)+\varphi_2)} + E^3(t, t - t_3) e^{i(\omega_3(t-t_3)+\varphi_3) + c.c.}. \tag{4}
\]

Here the envelopes \( E^l(r, t) \) are determined by the optimization procedure for localized pulses. The detected signal (selected quantum pathways of the full dipole density) is measured with heterodyne detection via phase cycling by repeating the experiment several times for different phases \( \varphi_1, \varphi_2, \) and \( \varphi_3 \) [Fig. 4(a)].

In general the polarization, created by three pulses applied to the quantum dots, is described by many quantum pathways in Liouville space. In the following way, we can extract a subset of the Liouville pathways by extracting a certain phase combination of \( \varphi_1, \varphi_2, \) and \( \varphi_3 \). The detected dipole density for different phases can be written as

\[
P(t, \varphi_1, \varphi_2, \varphi_3) = P(t, \varphi_1, \varphi_2, \varphi_3) + c.c., \tag{5}
\]

\[
P(t, \varphi_1, \varphi_2, \varphi_3) = \sum_{lmn} c_{123,lmn} P_{lmn}(t),
\]

with \( c_{123,lmn} = e^{i(\varphi_1 + \varphi_2 + \varphi_3)} \) for \( m+n+|m| = 1 \) or \( 3 \) for resonant excitation and \( P_{lmn}(t) \) being the part of the detected polarization with phase dependence \( \varphi_1 + \varphi_2 + \varphi_3 \). \( c_{123,lmn} \) can be viewed as a matrix with first index \( \varphi_3 \), second index \( l,m,n \), and \( |l| + |m| + |n| = 1 \) or 3 for resonant excitation and \( P_{lmn}(t) \) being the part of the detected polarization with phase dependence \( \varphi_1 + \varphi_2 + \varphi_3 \). So that the matrix \( c_{123,lmn} \) is invertible, we can extract the signal with a specific phase combination \( \varphi_3 = l \varphi_1 + m \varphi_2 + n \varphi_3 \) (selecting particular pathways) using \( P_{lmn}(t) = \sum_{l,m,n} c_{123,lmn} P(t, \varphi_1, \varphi_2, \varphi_3) \). Details of this phase-cycling
procedure can be found in Ref. 41. Typical examples for such signals are the photon echo \( \varphi_4 = \varphi_1 + \varphi_2 + \varphi_3 \) and antiphoton echo \( \varphi_2 = \varphi_1 - \varphi_2 + \varphi_3 \) (cf. Ref. 8).

### C. Double-quantum-coherence signal

We focus on the double-quantum-coherence signal, a third-order signal with the contributing phase combinations \( \varphi_1 = \varphi_1 + \varphi_2 - \varphi_3 \). In the case of a system where the ground state, single-exciton state, and two-exciton state form three bands [Fig. 1(b)], only the two Liouville pathways depicted in Fig. 4(b) will contribute to the signal with \( \varphi_4 = \varphi_1 + \varphi_2 - \varphi_3 \).

In the case of the three-band model [Fig. 1(b)] only two Liouville pathways can contribute. The part of the polarization attributed to \( \varphi_4 \), i.e., \( P^{(3)}_{1,1,1} (t) \), which depends on the delay times can be written using a response function:

\[
P^{(3)}_{1,1,1} (t) = \int_{0}^{\infty} dt_{1} \int_{0}^{\infty} dt_{2} \int_{0}^{\infty} dt_{3} \times R^{(3)}_{1,1,1} (t,t_{1},t_{2},t_{3})
\]

Note that we include the optical fields in the definition of the response, which is rather uncommon, but for the use of localized fields this notation will simplify the discussion. The response function \( R^{(3)}_{1,1,1} \) can be divided into the contributions of two Liouville pathways, extracted from the full response function:

\[
R^{(3)}_{1,1,1}(t,t_{1},t_{2},t_{3}) = \left( \frac{i}{\hbar} \right)^{3} \text{tr} \left[ \mu_{1} G (t_{1}) \mu_{2} G (t_{2}) \mu_{3} G (t_{3}) \rho_{0} \right]
\]

Here, the electron electric-field interaction Liouvillian \( H_{el-L} \) and \( \varphi_4 = \varphi_1 + \varphi_2 + \varphi_3 \) can, Green’s function \( G (t) \) with \( \rho (t) \rho (0) = \theta (t) \exp (-\frac{1}{\hbar} H_{el-L} t) \rho (0) \exp (-\frac{1}{\hbar} H_{el-L} t) \), and the dipole operator \( \mu = \sum_{ij} \mu_{ij} \langle \hat{g}^{\dagger} \rangle (\hat{l}_{i}) + \text{h.a.} \) For our excitonic three-band system, for the far-field excitation we insert the light-matter Hamiltonian in local basis:

\[
H_{el-L} = \sum_{i} \mu_{gi} \cdot E (t) |g\rangle \langle i| + \sum_{ij} \mu_{gi} \cdot E (t) |j\rangle \langle i| + \text{H.a.}
\]

The Hamilton operator can also be reformulated in the delocalized basis:

\[
H_{el-L} = \sum_{e} \mu_{ge} \cdot E (t) |e\rangle \langle e| + \sum_{ef} \mu_{ef} \cdot E (t) |e\rangle \langle f| + \text{H.a.}
\]

with the delocalized exciton dipole matrix elements \( \mu_{ge} = \sum_{i} c_{i}^{*} \mu_{gi} \) and \( \mu_{ef} = \sum_{ij} c_{i}^{*} \mu_{gi} c_{j} \). We insert Eq. (9) into Eq. (7) and collect for \( R^{(3)}_{1,1,1} \) only the terms proportional to \( \exp (t (-\varphi_3 + \varphi_2 + \varphi_1)) \) and end up with the response from two contributing Liouville pathways [Fig. 4(b)] assuming no temporal pulse overlap:

\[
R^{(3)}_{1,1,1}(t,t_{1},t_{2},t_{3}) = \left( \frac{i}{\hbar} \right)^{3} e^{\text{ia}(t_{1}+t_{2}+t_{3})} \times \mu_{ge} \cdot E^{3} (t_{3} - t_{1})
\]

\[
\times \mu_{ge} \cdot E^{2} (t_{2} - t_{1})
\]

\[
\times \mu_{ef} \cdot E^{2} (t_{2} - t_{1})
\]

\[
\times e^{-i\xi_{e}(t_{1})} \times e^{-i\xi_{f}(t_{2})} \times e^{-i\xi_{g}(t_{3})}.
\]

Here, \( \xi_{nm} = \omega_{nm} - i \gamma_{nm} \), with \( \omega_{nm} = \omega_{n} + \omega_{m} \) including the exciton frequencies \( \omega_{n} \) and the dephasing and relaxation rate \( \gamma_{nm} \) for a Lorentzian dephasing model.

In both pathways i and ii, we have a coherence between the single-exciton and ground states in between the first and second pulses and a coherence between the two-exciton to ground states in between the second and third pulses. After the third pulse the system is either in a single-exciton coherence (pathway ii) or ground-state to two-exciton coherence (pathway i) or ground-state to single-exciton coherence (pathway ii). We consider for further analysis the heterodyne detected signal, where the emitted signal \( P^{(3)}_{1,1,1} (t) \) is mixed with the field of a local oscillator \( E_{r} \):

\[
S_{3_{111}}^{(3)} (t_{1},t_{2},t_{3}) = \int_{-\infty}^{\infty} dt \cdot P^{(3)}_{1,1,1} (t) \cdot E^{3} (t)e^{ia_{t}}.
\]

\( S_{3_{111}}^{(3)} (t_{1},t_{2},t_{3}) \) is a complex quantity. A measurement obtains the real part of \( S_{3_{111}}^{(3)} (t_{1},t_{2},t_{3}) \). However, the use of a local oscillator in heterodyne detection allows—by twisting its
for the delocalized states. We see that no delocalized dipole moments are formed, since the effective response depends on the spatial distribution of the electric field.

Using far-field excitation for pulses $E^2$ and $E^3$, the local oscillator $E^4$ for heterodyne detection, and a localized excitation for the first pulse $E^1$ at dot $i$ ($E^1 \rightarrow E^1_\text{i}$), the double-quantum-coherence signal $S_{k_{3i}}(i, \Omega_1, \Omega_2, t_3) = S_{k_i}^{(3)}(i, \Omega_1, \Omega_2, t_3) + S_{k_i}^{(3)}(i, \Omega_1, \Omega_2, t_3)$ now depends on the chosen quantum dot $i$ and reads

$$S_{k_{3i}}^{(3)}(i, \Omega_1, \Omega_2, t_3) = S_{k_i}^{(3)}(i, \Omega_1, \Omega_2, t_3) + S_{k_i}^{(3)}(i, \Omega_1, \Omega_2, t_3)$$

(E. Discussion of the double-quantum-coherence signal)

Figure 5 shows the far-field double-quantum-coherence signal $[E^{(1)}_i(r_j, \omega) \approx E^{(1)}(\omega)]$, cf. Sec. III C absolute value [Fig. 4(a)] and imaginary value [Fig. 4(b)]. The frequency of the single-to-exciton to ground-state coherence can be seen on the $\Omega_1$ axis and the frequency of the two-exciton to ground-state coherence on the $\Omega_2$ axis. Clearly, for the far-field excitation in Figs. 5(a) and 5(b) we see resonances connecting to coherence of several states $e$ and $f$. If we select a frequency $\Omega_1 = \omega_{ef}$, we see along the $\Omega_2$ axis which specific two-exciton states are connected via dipole moments to the single-exciton state $e_j$ and vice versa. A comparison of the dipole moments connected to two different peaks works only roughly, since two Liouville paths interfere and the degree of destructive interference is different for every peak.

A dominant peak (A) in the absolute value spectrum [Fig. 5(a)] is connected to $e_2$ and $f_2$, a second strong peak is connected to $e_2$ and $f_1$, and some further peaks with smaller oscillator strength can be seen at a lower single-exciton energy ($e_3$ and $f_2$,$e_3$ and $f_1$), $e_1$ and $e_2$ are well resolved, and $e_3$ shows up as a spectral shoulder. This shows that the system has three single-exciton and three two-exciton states.

Figure 5(c) shows the signal with the first pulse localized at either quantum dots 1, 2, or 3. The localization of the first pulse...
gives information about the single-exciton states contributing to the ground-state–single-exciton transition occurring during the first pulse. Localization at quantum dot (QD) 1 shows that all resonances connected to the delocalized exciton state \( e_1 \) disappear. Overall, this shows that quantum dot 1 contributes strongly only to the formation of single-exciton states \( e_2 \) and \( e_3 \) but not to the buildup of \( e_1 \). Similar information is obtained for excitation of quantum dots 2 and 3 [see Fig. 5(c)]. For example, the exciton state \( e_2 \) is formed by quantum dots 1 and 2. Another interesting feature is the peak connecting \( e_3 \) and \( f_1 \). This peak is only visible at the localized spectrum at QD 2 and QD 3 and not in the far-field spectrum. This is caused by the fact that \( e_3 \) is an antisymmetric delocalized state between QDs 2 and 3, seen by the opposite sign of the peak in the QD 2 and QD 3 spectrum. For far-field excitation, these two antiparallel dipoles interfere destructively, so that the resonance is not observed.

We next use the localized double quantum coherence to extract the wave function coefficients \( c_i^e \) and therefore all quantum dot interactions.

**IV. EXTRACTING THE SINGLE-EXCITON WAVE FUNCTION**

All ingredients are now available to extract the single-exciton wave function. We start from the localized signal in Eqs. (20)–(22) and see that the sum over \( e \) and \( j \) prevents us from extracting a particular coefficient \( c_i^e \). Assuming ideal localization of the first pulse at a particular quantum dot \( i \) \([E_i^j(r,\omega_{eg}) \approx \delta_{ij} E_i^j(r,\omega_{eg})]\) removes the sum over \( j \) in Eqs. (20)–(22). Of course, any deviation from ideal localization will result in an error in the measurement of the coefficients (see below).

For removing the sum over \( e \) and selecting a particular single-exciton state \( e \), we choose the frequencies \( \Omega_1 = \Omega_1^e \) and \( \Omega_2 = \Omega_2^e \) in a way that only a specific peak caused by single-exciton to ground state \( \omega_{eg} \) and two-exciton to ground state coherences \( \omega_{fg} \) connected to \( e \) contributes, as suggested by the denominators in Eqs. (20)–(22). Again, if peaks for different single-exciton states overlap, errors are introduced to the reconstruction. However, two-dimensional spectroscopy has less spectral overlap than one-dimensional spectroscopy, since the peaks are separated by an additional degree of freedom: the additional frequency axis.) This yields

\[
S_{kii}^{(3)}(i,\Omega_1^e,\Omega_2^e,\tau_3) = S_{kii}^{(3)}(i,\Omega_1^e,\Omega_2^e,\tau_3) + S_{kii}^{(3)}(i,\Omega_1^e,\Omega_2^e,\tau_3),
\]

(23)

\[
S_{kii}^{(3)}(i,\Omega_1^e,\Omega_2^e,\tau_3) \approx \frac{1}{\hbar^2} \sum_{c,f} \mu_{c'f'} \cdot E^{4\times}(\omega_{fg})\mu_{c'f'} \cdot E^{4\times}(\omega_{fg}) \times \mu_{c'f'} \cdot E^{2}(\omega_{fg})c_i^e \mu_{c'f'} \cdot E_{1}^{j}(r,\omega_{eg}) \times \exp(-i\xi_{c'f'}\tau_3) / (\Omega_2 - \xi_{fg})(\Omega_1 - \xi_{eg}),
\]

(24)

\[
S_{kii}^{(3)}(i,\Omega_1^e,\Omega_2^e,\tau_3) \approx -\frac{1}{\hbar^2} \sum_{c,f} \mu_{c'f'} \cdot E^{4\times}(\omega_{fg})\mu_{c'f'} \cdot E^{4\times}(\omega_{fg}) \times \mu_{c'f'} \cdot E^{2}(\omega_{fg})c_i^e \mu_{c'f'} \cdot E_{1}^{j}(r,\omega_{eg}) \times \exp(-i\xi_{c'f'}\tau_3) / (\Omega_2 - \xi_{fg})(\Omega_1 - \xi_{eg}).
\]

(25)

We see that here the double-quantum-coherence signal is proportional to \( c_i^e \mu_{c'f'} \cdot E_{1}^{j}(r,\omega_{eg}) \), that is, to the strength \( c_i^e \) the \( j \)th quantum dot contributes to the delocalized wave function. This fact is used to develop a scheme to extract the coefficients \( c_i^e \) from measured data:

As input information, the dipole moments \( \mu_{gi} \) of the individual uncoupled quantum dots are required, and the dipole moments can be measured or calculated. As measurement, carry out the localized double-quantum-coherence signal \( S_{kii}^{(3)}(i,\Omega_1^e,\Omega_2^e,\tau_3) \), for a localization on all quantum dots \( i \). If the field strength and polarization direction are different for localization at different quantum dots, we need to obtain the electric field along the local dipole \( \mu_{gi} \cdot E_{1}^{j}(r,\omega_{eg}) \).

Now, we select the excitonic state \( e \equiv \epsilon_{eg}, \) whose coefficients \( c_i^e \) should be extracted. We determine the the frequencies \( \Omega_1 \approx \omega_{eg}, \Omega_2 \approx \omega_{fg} \) showing a strong correlation to \( \epsilon_{eg} \) using the double-quantum-coherence signal without spatial localization. Now in the postprocessing of the data, we use that \( c_i^{e*} \propto S_{kii}^{(3)}(i,\Omega_1^e,\Omega_2^e,\tau_3) / [\mu_{gi} \cdot E_{1}^{j}(r,\omega_{eg})] \) at the positions \( \Omega_1 \approx \omega_{eg}, \Omega_2 \approx \omega_{fg} \) [Eqs. (23)–(25)]. \( c_i^{e*} \) can now be determined up to an proportionality factor \( A: c_i^{e*} = S_{kii}^{(3)}(i,\Omega_1^e,\Omega_2^e,\tau_3) / [\mu_{gi} \cdot E_{1}^{j}(r,\omega_{eg})A] \) for every quantum dot \( i \), using the same frequencies \( \Omega_1, \Omega_2, \Omega_3 \). Since the wave function is normalized, \( |A|^2 = \sum_i |A_i c_i^{e*}|^2 \) holds. We thus get \( A \) up to a global phase and set \( A = |A| \). We obtain \( c_i^{e*} = S_{kii}^{(3)}(i,\Omega_1^e,\Omega_2^e,\tau_3) / [\mu_{gi} \cdot E_{1}^{j}(r,\omega_{eg})A] \). This gives the delocalized wave function \( |\epsilon_{eg}| = \sum_i c_i^{e*} |i| \).

Note that these steps constitutes a quantum state tomography. The local basis is uniquely determined up to an arbitrary phase for every quantum dot: The expansion coefficient \( c_i^e \) depends on that choice.
To demonstrate the success of the tomography, we compare in Fig. 6 the elements of the reconstructed wave function for the strongest contribution, that is, state \( e_2 \) [marked with A in Fig. 5(a)], to the original wave function resulting from the input parameters in the Hamiltonian. The agreement for both the amplitude and the relative phase is quite good. The difference results from a nonperfect localization \( E^i_j(r_j, \omega_{eg}) \neq \delta_j E^i_j(r_i, \omega_{eg}) \) resulting from realistic Maxwell simulation from Sec. IIIA. This error is marked by the error bars in Fig. 6. It is caused by a weak excitation of quantum dots, which a ideally localized pulse should not excite. Such a nonideal excitation leads to cross talk between the coefficients. The error bars are estimated to be smaller than \( \Delta \approx \sum modes \neq 3|E_i(t_j)|/|E_i(t_i)| \).

Note that in general the procedure works also for other methods than heterodyne detection in the far field, including a localized detection of polarization or fluorescence, as long as the detection is the same for a localization of the first pulse at different quantum dots. The only limitation is that the phase of the coefficients can be detected only with methods that can measure complex signals. For other types of detection, like homodyne detection, we can also extract the absolute value of the coefficients but not their phase.

V. FILTERING COHERENT SPECTRA

As additional useful application, we show that strong, undesired resonances can be selectively suppressed from coherent spectra. This can be advantageous while investigating weak resonances that are masked by other strong resonances: Often, it is not clear whether weak resonances constitute a vibrational side peak connected to a dominating strong excitonic peak or a different, much weaker excitonic resonance. This can also be solved by selectively removing excitonic resonances from measured spectra and applying a filter algorithm.

As input information for the filter algorithm, we have to determine expansion coefficients \( c^{e_\alpha}_i \) for the specific state \( e_\alpha \) for all quantum dots \( i \) whose contributions we want to filter out. Additionally, we need all dipole moments \( \mu^{e_\alpha}_{gi} \) of the individual nanostructure and also the electric field along the local dipole \( \mu^{e_\alpha}_{gi} \cdot E_i^i(r_i, \omega_{eg}) \).

As measurement we record a localized version of the spectrum to be filtered. The localized pulse should excite a ground-state to single-exciton transition for all quantum-dot positions. For the double quantum coherence, this will be the signal \( S_{i(\Omega_1, \Omega_2, t_3)}(i, \Omega_1, \Omega_2, t_3) \) for every quantum dot \( i \).

For postprocessing we discuss the expression

\[
S_{i(\Omega_1, \Omega_2, t_3)} = S_{i(\Omega_1, \Omega_2, t_3)} - \sum_i c^{e_\alpha}_i \cdot \mu^{e_\alpha}_{gi} \cdot E_i^i(r_i, \omega_{eg}) f_{e_\alpha}(\Omega_1, \Omega_2, t_3) = \sum_i c^{e_\alpha}_i \mu^{e_\alpha}_{gi} \cdot E_i^i(\omega_{eg}) f_{e_\alpha}(\Omega_1, \Omega_2, t_3),
\]

which gives a spectrum where all contribution of \( e_\alpha \) during the first pulse are filtered out.\(^{51}\)

The single-exciton peak \( e_2 (\alpha = 2) \) dominating the spectrum in Fig. 5(a) is filtered out in Fig. 5(d). This spectrum reveals information about states initially covered by the dominant contribution of \( e_2 \). The procedure can be applied iteratively, using the filtered spectra for obtaining the other excitonic states. This can enhance the reconstruction of the exciton states.

The filtering method can also be applied to other spectroscopic signals as long as a phase-sensitive detection is used and a localized signal, whose contributions are proportional to the single-exciton expansion coefficients, can be measured.

VI. CONCLUSION AND OUTLOOK

The presented quantum state tomography for the extraction of the delocalized single-exciton wave function coefficients can also be applied to other impulsive two-dimensional spectra. The single-exciton to two-exciton transition in double quantum coherence using the localization of the second pulse also can be used to extract the two exciton coefficients. However, since this problem is more complex, it will be the subject of future work.

In conclusion, our simulations demonstrate a quantum state tomography that can be used to reconstruct individual wave functions of coupled emitters acting only collectively in the far field. In addition, localized excitations are useful to remove unwanted strong resonances to uncover weak or hidden excitonic resonances. All of these features are not accessible in standard far-field spectroscopy. Similar configurations can be alternatively achieved by applying four pulses and using phase cycling to detect a desired component,\(^{43}\) for example, with phase: \( \varphi = \varphi_1 + \varphi_2 - \varphi_3 - \varphi_4 \). We therefore believe that the proposed quantum state tomography opens a new path for the detection of many-body interactions on the nanoscale. The proposed protocol is more general as presented here, since fluorescence can also be used rather than heterodyne detection of optical fields.\(^{43}\)

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The idea behind the filtering algorithm is that we can write the localized spectrum in terms of individual contributions $f_{e'}$ caused by resonances for different single excitons $e'$, where $f_{e'}$ are calculated from measured spectra [cf. Eq. (27)]:

$$S_{III}(i, \Omega_1, \Omega_2, t_3) = \sum_{e'} c_i^{e'} \mu_{ig}^{\ast} \cdot E_1^{\ast}(r_i, \omega_{eg}) f_{e'}(\Omega_1, \Omega_2, t_3). \quad (28)$$

Multiplying the equation with $c_i^{e'}$ and summing over $i$ yields a scalar product, and we get $f_{e'}$ defined using the localized spectra [Eq. (27)]. The far-field double-quantum-coherence spectrum can also be calculated using $f_{e'}$:

$$S_{III}(\Omega_1, \Omega_2, t_3) = \sum_{e'} c_i^{e'} \mu_{ig}^{\ast} \cdot E_1^{\ast}(\omega_{eg}) f_{e'}(\Omega_1, \Omega_2, t_3). \quad (29)$$

It is expressed using summands for every contributing single exciton $e'$, and the summand of the exciton to be filtered can be subtracted. This is possible, since every summand can be calculated using $f_{e'}$, which can be calculated from the experimental data of the localized spectrum, if the expansion coefficients of the single-exciton wave function of $e'$ are known.