Dynamics of excitons in individual InAs quantum dots revealed in four-wave mixing spectroscopy: supplementary material

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1. SAMPLE PREPARATION AND CHARACTERIZATION

The MBE grown sample contains a layer of annealed and capped InAs QDs with a nominal density of $2.2 \times 10^9$ cm$^{-2}$. They are embedded in an asymmetric GaAs/AlGaAs micro-cavity exhibiting a low quality factor $Q = 170$, resulting in a mode centered at 910-915 nm with a FWHM of around 10 nm. The femto-second laser pulse trains are spectrally matched with such a large spectral window and efficiently penetrate into the structure. Furthermore, the intra-cavity field is enhanced by a factor of $\sqrt{Q} = 13$ improving the coupling between $E_{1,2,3}$ and the electric dipole moment $\mu$ of the transition. Thus, the resonant field required to drive the FWM is reduced by a factor $Q^{3/2} \approx 2200$ and the signal-to-noise ratio of the interferometrically detected FWM is amplified accordingly.

The sample is intentionally doped with Si ($\delta$-doping with a nominal density of $1.8 \times 10^{10}$ cm$^{-2}$; layer located 10 nm below the QD plane). To identify the spatial and spectral location of the QD transitions we perform hyperspectral imaging [5, 6]. In Fig. S1 a we present an example of such imaging performed in a confocal micro-photoluminescence (PL) experiment. Each bright spot corresponds to a QD emission, primarily attributed to recombination of negative trions (GX$^-$) due to the $n$-doping. We detect high PL counting rates on the order of $10^5$/sec at the QD saturation. Such an unusually bright PL emission is attributed to the presence of oval photonic defects on the sample surface [3, 4], acting as natural micro-lenses [7]. Additionally, the inhomogeneous broadening due to spectral wandering is largely reduced [1, 4] indicating an excellent structural quality of these QDs.

The FWM hyperspectral imaging at the same sample area and spectral range is shown in Fig. S1 b. The three QDs at $(x, y) \approx (-2 \mu m, -5 \mu m)$, $(2 \mu m, -5 \mu m)$ and $(7 \mu m, -4 \mu m)$ (marked with green boxes in Fig. S1) exhibit both PL and FWM signals and were used to align the figures. However, other QDs show different distribution of the peak heights in FWM as compared to the PL. This is expected from the different properties determining the signal strength in both measurements: in FWM the dipole moment is probed, while in PL generally the more complex phonon-assisted carrier relaxation combined with a capture of the exciton also lead to a signal. To demonstrate the high spectral and spatial selectivity of the FWM compared to the PL, Figs. S1 c and d compare both PL and FWM signals and were used to align the figures. However, other QDs show different distribution of the peak heights in FWM as compared to the PL. This is expected from the different properties determining the signal strength in both measurements: in FWM the dipole moment is probed, while in PL generally the more complex phonon-assisted carrier relaxation combined with a capture of the exciton also lead to a signal. To demonstrate the high spectral and spatial selectivity of the FWM compared to the PL, Figs. S1 c and d compare both PL and FWM obtained from the same sample spot, defined by the diffraction limited size (0.7 $\mu$m) of the excitation laser. In Fig. S1 c we show a neutral exciton complex, which is only present in few % of the QDs. The exciton-biexciton system is straightforwardly recognized...
The light field coupling

\[
M = \begin{pmatrix}
0 & \Omega_{\nu^+}^* & \Omega_{\nu^-} & 0 \\
\Omega_{\nu^+} & 0 & 0 & \Omega_{\nu^-}^* \\
\Omega_{\nu^-} & 0 & 0 & \Omega_{\nu^+}^* \\
0 & \Omega_{\nu^+} & \Omega_{\nu^-} & 0
\end{pmatrix}
\]  

(4)

describes the allowed transitions via the Rabi frequencies

\[
\Omega_{\nu^\pm} = \sum_j M_0 j |e_{\nu^\pm}|. 
\]

(5)

\(M_0\) is the bulk dipole matrix element and \(e_{\nu^\pm}\) the polarization vector. The polarization of the system is given by

\[ p = M_0 (|0\rangle \langle \sigma^+ | + |\sigma^- \rangle \langle B|) e_{\sigma^-} 
+ M_0 (|0\rangle \langle \sigma^- | + |\sigma^+ \rangle \langle B|) e_{\sigma^+}. \]

By the diagonalization the degeneracy of the single excitons is lifted and the exciton energies are \(\hbar \omega_X = \hbar \omega_\nu - \delta/2\) and \(\hbar \omega_Y = \hbar \omega_\nu + \delta/2\). The light field coupling changes to

\[
M = \begin{pmatrix}
0 & \Omega_X^* & \Omega_Y & 0 \\
\Omega_X & 0 & 0 & \Omega_Y^* \\
\Omega_Y & 0 & 0 & \Omega_X^* \\
0 & \Omega_X & \Omega_Y & 0
\end{pmatrix}
\]  

(8)

with

\[
\Omega_X = \frac{1}{\sqrt{2}} (\Omega_{\nu^+} + \Omega_{\nu^-}), \quad \Omega_Y = \frac{i}{\sqrt{2}} (\Omega_{\nu^+} - \Omega_{\nu^-}).
\]

The time evolution of the density matrix \(\rho\) is calculated assuming a sum of \(\delta\)-pulses yielding the Rabi frequencies for circular polarization

\[
\Omega_{\nu^\pm} = \sum_j \frac{\theta_{\nu^\pm}}{\tau} e^{i \varphi_{\nu^\pm}} \delta(t - t_j)
\]

(9)

with arrival times \(t_j\), pulse areas \(\theta_{\nu^\pm}\) and phases \(\varphi_{\nu^\pm}\). For a pulse sequence with linear polarizations \(\alpha_j\) with respect to \(X\) and pulse areas \(\theta_j\) the Rabi frequencies read

\[
\Omega_X = \sum_j \sqrt{2} \theta_j e^{i \phi_j} \cos(\alpha_j),
\]

\[
\Omega_Y = \sum_j -\sqrt{2} \theta_j e^{i \phi_j} \sin(\alpha_j).
\]

In the case of \(\delta\)-pulses the time evolution of the system can be calculated by matrix multiplication \[8\]. In between the pulses the dynamics is given by

\[
\rho_{\nu^\nu'}(t) = \rho_{\nu^\nu'}(0) e^{i \Lambda_{\nu^\nu'}(t)}
\]

(10)
which corresponds to an energetic broadening of wandering of individual transitions. This phenomenon induces with (i.e. up to several homogeneous linewidths) can be included in the calculations by multiplying the FWM-polarization with a Gaussian function \[10\] as follows:

\[
\rho(\nu) = \rho_{BB}(0) e^{\gamma t}.
\]

\[
\rho(\nu) = \rho_{XX}(0) + \rho_{BB}(0) (1 - e^{-\gamma t}) e^{-\gamma t}.
\]

\[
\rho_{YY}(t) = \rho_{YY}(0) + \rho_{BB}(0) (1 - e^{-\gamma t}) e^{-\gamma t}.
\]

\[
\rho_{GC}(t) = 1 - \rho_{XX}(0) + \rho_{YY}(0) + \rho_{BB}(0) (2 - e^{-\gamma t}) e^{-\gamma t}.
\]

The time \( t = 0 \) corresponds to the time directly after each pulse.

>From this, we can calculate the dynamics of all elements of the density matrix:

\[
\rho_{XX}(t) = \rho_{XX}(0) e^{\gamma t}.
\]

\[
\rho_{YY}(t) = \rho_{YY}(0) + \rho_{BB}(0) (1 - e^{-\gamma t}) e^{-\gamma t}.
\]

\[
\rho_{GC}(t) = 1 - \rho_{XX}(0) + \rho_{YY}(0) + \rho_{BB}(0) (2 - e^{-\gamma t}) e^{-\gamma t}.
\]

The phase dependence of the polarization. In general, all polarizations have parts depending on different orders and combinations of the phases \( \psi_i \) of the pulses. The two-pulse FWM for coherence dynamics is given by the phase combination \( \phi_{12} \) which modulates the heterodyning at \( \Omega_2 - \Omega_1 \) and \( \Omega_3 - \Omega_2 - \Omega_1 \). This identifies the polarization of the FWM signal indicated by \( p_{FWM} \).

For the sake of simplicity, in the case of population dynamics we use \( \tau_{12} = 0 \) ps to mimic the short time decay before the first two pulses. From the polarization the FWM signal \( S_{\nu\nu} \) is obtained by a Fourier transform at the selected frequency

\[
S_{\nu\nu} = \int_0^\infty p_{FWM} e^{i\omega t} dt |_{\omega = \omega_{\nu} - \omega_{\nu'}}.
\]

If the polarization \( a \) is not along one axis of the QD, the signals are added according to the angle of the heterodyning (reference) beam \( \alpha \) with \( S_{XY} = \cos^2(\alpha) S_{XX} + \sin^2(\alpha) S_{YY} \).

In the FWM signal, charge fluctuations can play an important role leading to an inhomogeneous broadening via spectral wandering of individual transitions. This phenomenon induces a photon echo in FWM transients of single QDs, when probing the coherence \(9-11\). The residual inhomogeneous broadening (i.e. up to several homogeneous linewidths) can be included in the calculations by multiplying the FWM-polarization with a Gaussian function \(10\) as follows:

\[
p_{FWM} \rightarrow p_{FWM} e^{-\left(\frac{(\omega - \omega_0)^2}{2\sigma^2}\right)}
\]

For most cases the inhomogeneous broadening can be neglected. We only included it to model the data in Fig. 4 with \( \sigma = 67 \) ps, which corresponds to an energetic broadening of \( h\sigma \approx 10 \) µeV.

REFERENCES


