Supplementary Material

This document intends to provide supplementary information to the article *Coherence and degree of time-bin entanglement from quantum dots* by T. Huber, L. Ostermann, M. Prilmüller, G. S. Solomon, H. Ritsch, G. Weihs and A. Predojević.
The quantum dot sample was held at a temperature of 4.8 K. It contained low density self-assembled InAs quantum dots embedded in a planar micro-cavity, increasing the vertical collection of photons. The excitation light was derived from a tunable 82 MHz repetition rate Ti:Sapphire pulsed laser.
To give a comparison to time-bin entanglement results obtained using parametric down-conversion we measured visibilities in three orthogonal bases. For the state generated using 12 ps pulses we find the visibilities of 94(2) % in the classically correlated basis (time basis) and 74(5) % and 67(5) % in the bases that indicate entanglement (energy bases).

THEORETICAL MODEL

The quantum dot system having a ground state $|g\rangle$, an intermediate exciton state $|x\rangle$ and a biexciton state $|b\rangle$ is described by the Hamiltonian

$$H = \frac{1}{2}\left(\Omega_1 |g\rangle\langle x| + h.c.\right) + \frac{1}{2}\left(\Omega_2 |x\rangle\langle b| + h.c.\right) + \left(\Delta_x - \Delta_b\right)|x\rangle\langle x| - 2\Delta_b |b\rangle\langle b|\,, \tag{1}$$

where Ω_1 and Ω_2 are the (identical) Rabi frequencies emerging from the coupling of the ground and exciton and the exciton and biexciton state, respectively, by the excitation laser pulse. Here, Δ_x is the difference between the virtual two-photon level and the exciton state of the quantum dot, while Δ_b is the detuning between the virtual two-photon level and the laser driving the quantum dot. In our experiment, we drive the two-photon virtual level resonantly, therefore $\Delta_b = 0$. Δ_x has been measured to be $\Delta_x = 2\pi \cdot 0.335\text{THz}$.

Our quantum dot system is subject to dissipative processes, therefore, we introduce the definition of a dissipator for the operator $\hat{A}$ as

$$D(\hat{A}) := 2\hat{A}^\dagger \rho \hat{A} - \hat{A}\hat{A}^\dagger \rho - \rho \hat{A}\hat{A}^\dagger. \tag{2}$$

With this definition we can now easily write down the loss processes in the system, which are

$$\begin{aligned}
\mathcal{L}_1 &= \frac{\gamma_b}{2} D\left(|b\rangle\langle x|\right) && (3)\\
\mathcal{L}_2 &= \frac{\gamma_x}{2} D\left(|x\rangle\langle g|\right) && (4)\\
\mathcal{L}_3 &= \frac{\gamma_b^{\text{deph}}}{2} D\left(|b\rangle\langle b| - |x\rangle\langle x|\right) && (5)\\
\mathcal{L}_4 &= \frac{\gamma_x^{\text{deph}}}{2} D\left(|x\rangle\langle x| - |g\rangle\langle g|\right), && (6)
\end{aligned}$$

where the first two expressions consider population loss by spontaneous emission with the rates γ_b for the biexciton and γ_x for the exciton. These rates were measured to be $\gamma_b = 1/771\text{THz}$ and $\gamma_x = 1/405\text{THz}$. On the other hand, $\mathcal{L}_3$ and $\mathcal{L}_4$ account for the dephasing between the biexciton and exciton and the exciton and ground state, respectively. The rates have been assumed equal and are of the form

$$\gamma_b^{\text{deph}} = \gamma_x^{\text{deph}} = \gamma_{I_0} \cdot \Omega^{n_p} \tag{7}$$

where γ_{I_0} is the amplitude of the intensity-dependent dephasing and n_p represents the exponent of the intensity dependence. In our study we investigated $n_p = 2$ and $n_p = 4$, hence the linear ($n_p = 2$) and the quadratic ($n_p = 4$) intensity dependence of the dephasing. The value of γ_{I_0} has been obtained by fitting the model to the experimental data.

The full Liouvillian is hence

$$\mathcal{L} = \mathcal{L}_1 + \mathcal{L}_2 + \mathcal{L}_3 + \mathcal{L}_4 \tag{8}$$

and the time evolution of our system is governed by the master equation

$$\dot{\rho} = i\,[\rho, H] + \mathcal{L}. \tag{9}$$

The coherent excitation is described by time-dependent Rabi frequencies that for a Gaussian laser pulse envelope follow

$$\Omega_1 = \Omega_2 = \Omega \cdot \exp\left(-\frac{2\ln(2)\,(t-t_i)^2}{\sigma^2}\right), \tag{10}$$

where σ is the pulse length. In the experiment the pulse length can be varied from 4 ps to 20 ps. t_i is the center of the Gaussian pulse in the time domain.

Rabi oscillations

In order to gain some insight into the dynamics of our system we plot the normalized emission probabilities

$$P_b(t_f) \;=\; \gamma_b \int_0^{t_f} \langle b|\,\rho(t')\,|b\rangle\;\mathrm{d}t' \tag{11}$$

$$P_x(t_f) \;=\; \gamma_x \int_0^{t_f} \langle x|\,\rho(t')\,|x\rangle\;\mathrm{d}t' \tag{12}$$

as a function of $\Omega^2\sigma$, which is proportional to the energy per pulse (Fig. 1). Here, t_f is the time at which the photon is detected. For the constant dephasing the numbers $\gamma_x^{\mathrm{deph}} = 1/119\mathrm{THz}$ and $\gamma_b^{\mathrm{deph}} = 1/211\mathrm{THz}$ were used. These specific numbers are chosen in congruence with the $g^{(1)}(\tau)$ field correlation measurements performed on exciton and biexciton photons, respectively.

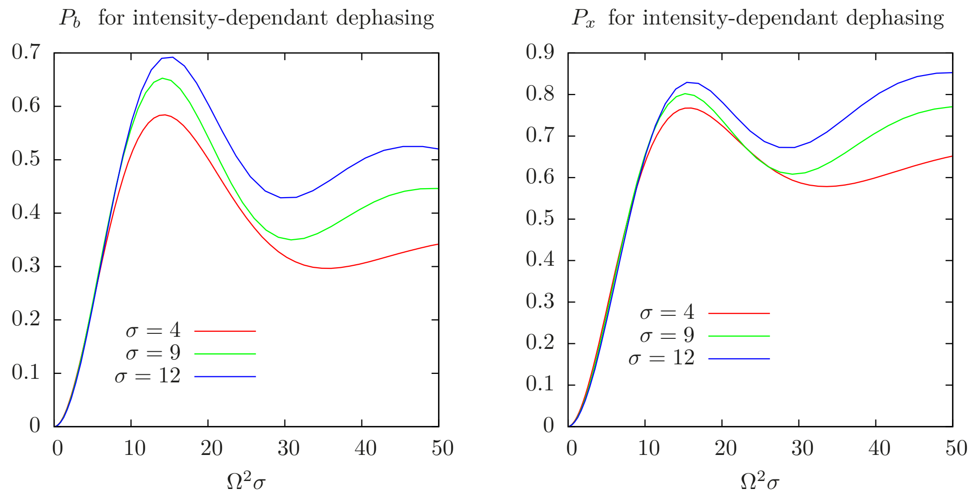

FIG. 1: Rabi oscillations for linearly intensity-dependent dephasing with an amplitude of $\gamma_{I_0} = 0.05$ as a function of the deposited energy. The emission probability from the biexciton (left) and the exciton (right) level is shown. Comparing this figure with the Figure 2 from the main text becomes clear that the ratio between the first Rabi maximum and its first minimum is largely affected by γ_{I_0}. The figure for $n_p = 4$ looks fairly similar and is therefore omitted.

As also explained in the manuscript, the population loss does not necessary need to originate exclusively from constant dephasing [1]. If we introduce the intensity-dependent dephasing, given by Eq. 8 in the manuscript, such that $\gamma_{I_0} = 0.05$ for $n_p = 2$ and $n_p = 4$, we obtain the normalized emission probabilities depicted in Fig. 1.

Population at low excitation power

Using second order time-dependent perturbation theory we can calculate the ratio between P_b and P_x for small driving strengths Ω. For this we will consider only the coherent part of the time evolution and neglect damping and dephasing.

Our Hamiltonian therefore will have the following form

$$H = H_0 + V = \begin{pmatrix} 0 & 0 & 0 \\ 0 & \Delta & 0 \\ 0 & 0 & 0 \end{pmatrix} + \begin{pmatrix} 0 & \Omega & 0 \\ \Omega & 0 & \Omega \\ 0 & \Omega & 0 \end{pmatrix}.$$

The formal solution of the Schrödinger equation in the interaction picture for a time-dependent perturbation V in the basis $\{|n\rangle\}$ is

$$c_n(t) = c_n(0) - i \sum_k \int_0^t \mathrm{d}t' \langle n| V(t') |k\rangle \exp\left(-i(E_k - E_n)t'\right) c_k(t').$$

By recursively inserting this equation into itself we obtain the first-order

$$c_n(t) = c_n(0) - i \sum_k \int_0^t \mathrm{d}t' \langle n| V(t') |k\rangle \exp\left(-i(E_k - E_n)t'\right) c_k(0)$$

and the second order perturbative solution, i.e.

$$\begin{aligned} c_n(t) =& c_n(0) - i \sum_k \int_0^t \mathrm{d}t' \langle n| V(t') |k\rangle \exp\left(-i(E_k - E_n)t'\right) \\ &\cdot \left[c_k(0) - i \sum_{k'} \int_0^{t'} \mathrm{d}t'' \langle k| V(t'') |k'\rangle \exp\left(-i(E_{k'} - E_k)t''\right) c_{k'}(0) \right]. \end{aligned}$$

When multiplying this expression we use that $V(t)$ is time-independent, while $E_{ab} = E_a - E_b$. We obtain

$$\begin{aligned} c_n(t) =& c_n(0) + \sum_k \langle n| V |k\rangle c_k(0) \frac{\exp\left(-iE_{kn}t\right) - 1}{E_{kn}} \\ &+ \sum_{k,k'} \langle n| V |k\rangle \langle k| V |k'\rangle \frac{c_{k'}(0)}{E_{k'k}} \left(\frac{\exp\left(-iE_{k'n}t\right) - 1)}{E_{k'n}} - \frac{\exp\left(-iE_{kn}t\right) - 1)}{E_{kn}} \right). \end{aligned}$$

Our quantum dot system has three states $|g\rangle$, $|x\rangle$ and $|b\rangle$. This system is initially in the ground state, which means $c_g(0) = 1$. This condition together with the structure of our perturbation (ii.e. only coupling ground and exciton and exciton and biexciton states) immediately reduces the sums to exactly one single term. Hence,

$$\begin{aligned} c_x(t) =& \frac{\Omega}{\Delta} \sin\left(\frac{\Delta t}{2}\right) \exp\left(-i\frac{\Delta t}{2}\right) \\ c_b(t) =& \frac{\Omega^2}{\Delta} \left[\frac{\sin\left(\frac{\Delta t}{2}\right)\cos\left(\frac{\Delta t}{2}\right)}{\Delta} - i \left\{ \frac{\sin\left(\frac{\Delta t}{2}\right)^2}{\Delta} + t \right\} \right] \end{aligned}$$

and the emission probability ratio therefore yields

$$\frac{P_b}{P_x} = \frac{\gamma_b \int_0^t |c_b(t')|^2 \, \mathrm{d}t'}{\gamma_x \int_0^t |c_x(t')|^2 \, \mathrm{d}t'} \sim \frac{\frac{\Omega^4}{\Delta^5)}}{\frac{\Omega^2}{\Delta^3}} = \frac{\Omega^2}{\Delta^2},$$

which goes to zero for $\Omega \to 0$.

Detection probabilities for two pulse excitation

Up to a geometric factor, the field emitted by a single quantum emitter at a position $\vec{x}$ is proportional to its dipole operator $\hat{\sigma}$ evaluated at the proper retarded time, i.e.

$$\hat{E}^+(\vec{x}, t) \sim \vec{e}(\vec{x}) \cdot \hat{\sigma}(t - R/c), \tag{13}$$

where $\vec{e}(\vec{x})$ represents the geometrical emission distribution and R the optical path length between emitter and detector. In our case we have the two emission dipoles that correspond to the exciton transition $\hat{\sigma}_x = |g\rangle\langle x|$ and the biexciton transition $\hat{\sigma}_b = |x\rangle\langle b|$. If we assume that the frequencies of the biexciton and the exciton photon are sufficiently distinct to allow distinguishable detection at separate detectors, we can approximate the fields at the detectors by

$$\hat{E}_x^+(t) = \eta(\hat{\sigma_x}(t - R_x/c) + e^{i\phi_x}\hat{\sigma_x}(t - R_x/c - \tau)) \tag{14}$$
$$\hat{E}_b^+(t) = \eta(\hat{\sigma_b}(t - R_b/c) + e^{i\phi_{xx}}\hat{\sigma_b}(t - R_b/c - \tau)). \tag{15}$$

Here, η is a general detection efficiency subsuming geometry and the actual detector efficiency η_D. The angles ϕ_x and ϕ_{xx} are the individual phase shifts in the x and xx interferometers and τ is the delay time between the two pulses. Using these expressions and assuming a sufficient time delay between the two pulses, so that all excitations have decayed in the dot upon arrival of the second pulse, we can factorize some expectation values and obtain the following average count rates,

$$\begin{aligned} I_x(t) &= \left\langle \hat{E}_x^-(t)\hat{E}_x^+(t)\right\rangle = \eta^2\left\langle e^{-i\phi_x}(\sigma_x^\dagger(t+\tau)\sigma_x(t) + e^{i\phi_x}\sigma_x^\dagger(t)\sigma_x(t+\tau)) + P_x(t) + P_x(t+\tau)\right\rangle \qquad (16)\\ &\approx \eta^2\left(\cos(\phi_x)\,\rho_{gx}\rho_{xg} + \rho_{xx}\right)\\ I_b(t) &= \eta^2\left\langle \hat{E}_b^-(t)\hat{E}_b^+(t)\right\rangle \approx \eta^2\left(\cos(\phi_{xx})\rho_{xb}\rho_{bx} + \rho_{bb}\right). \qquad (17)\end{aligned}$$

The corresponding probabilities can be obtained from our above calculations by integration over the pulse duration. Note, that some phase dependence of the signal can survive from the interference of the Rayleigh component of the scattering. In a similar way, for the two photon coincidence count probabilities we find

$$P_{bx}(t) = \eta^4\left\langle \hat{E}_b^-(t)\hat{E}_x^-(t)\hat{E}_x^+(t)\hat{E}_b^+(t)\right\rangle = \eta^4\left(\cos(\phi_x+\phi_{xx})\rho_{gb}\rho_{bg} + (1+\rho_{xx})\rho_{bb} + 2\cos(\phi_{xx})\rho_{xb}\rho_{bx}\right) \tag{18}$$
$$P_{xb}(t) = \eta^4\left\langle \hat{E}_b^+(t)\hat{E}_x^-(t)\hat{E}_x^+(t)\hat{E}_b^+(t)\right\rangle = \eta^4\rho_{xx}\rho_{bb}, \tag{19}$$

where $P_{bx}(t)$ corresponds to detecting the biexciton photon before the exciton photon and $P_{xb}(t)$ is the other way round. The dominant contribution comes from the ground state - biexciton coherence, which is induced by the phase of the excitation pulses and depends on the sum phase of the two interferometers. This behaviour is in an close analogy to the results obtained in experiments using down-conversion by a nonlinear crystal. The quantities presented here have to be averaged at least over the detector response time. Note, that we can also get the probability for four simultaneous counts P_4 , which simply reads $P_4 \approx 16\eta^8\rho_{bb}^2$ giving the product probability of a biexciton generated by a single pulse. This quantity is independent of any phase settings and we cannot detect more than four photons at exactly the same time, independent of how strong a pump we employ, due to pairwise anti-bunching.
In Fig. 2 we depict the evolution of the populations and coherences of the density matrix for a Gaussian pulse as a function of the pulse duration in an ideal system without any dephasing. We see that the initially coherence follows the population until a gap opens and incoherent biexciton population builds up.

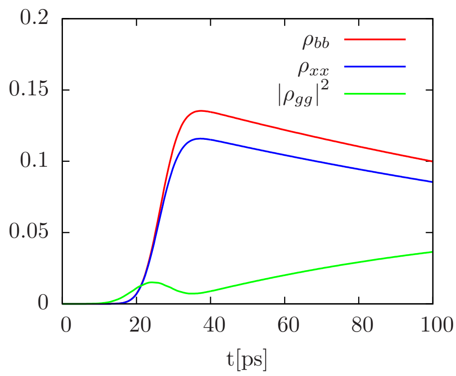

FIG. 2: Time evolution of exciton ρ_{xx} and biexciton ρ_{bb} population as well as the biexciton-ground coherence as a function of time for a Gaussian pulse of $\sigma = 12$ ps.

[1] Q. Q. Wang, et al. Physical Review B **72**, 035306 (2005).