

Resonant and antiresonant exciton-phonon coupling in quantum dot molecules

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Spins confined in optically active quantum dot molecules (QDMs) can be used for the deterministic generation of photonic graph states with tailored entanglement structures. Their usefulness for the generation of such nonclassical states of light is determined by orbital and spin decoherence mechanisms, particularly phonon-mediated processes dominant at energy scales up to a few millielectronvolts. Here, we directly measure the spectral function of orbital phonon relaxation between the energy states of the neutral exciton in a QDM and benchmark our findings against microscopic $\mathbf{k} \cdot \mathbf{p}$ theory. Our results reveal pronounced resonances and antiresonances in the phonon-relaxation rates, ranging from tens of μs^{-1} up to tens of ns^{-1} . Comparison with a kinetic model reveals the voltage (energy) dependent phonon coupling strength and fully explains the interplay between phonon-assisted relaxation and radiative recombination. The resonances and antiresonances enable further tunability of the exciton lifetime which can be leveraged to increase the lifetime of energetically unfavorable charge configurations needed for realizing efficient spin-photon interfaces and multidimensional cluster states.

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I. INTRODUCTION

Optically active double quantum dots forming a quantum dot molecule (QDM) have electrically tunable orbital structures [1–3] and can host two-spin states capable of creating one- and two-dimensional entanglement structures required in measurement based photonic quantum information processing [4–8]. Coherent spin-photon interfaces must be highly coherent and orbital spin dephasing is governed by interactions with the fluctuating nuclear spin environment and coupling to the lattice [9–12]. The main processes that limit the lifetime and coherence of excited orbital states in QDMs are spontaneous emission and phonon-induced orbital relaxation [13,14]. In most of the published works, these processes were treated as being additive [15] either via the impact of phonon scattering

in optical emission [16] or on coherent optical driving [17,18]. This approach is typically justified by the large difference between the picosecond timescales over which phonon-induced processes occur and nanosecond radiative lifetimes. The disparity between timescales does not necessarily hold in QDMs, since the interdot tunnel coupling and the field-tunable energy between excitons of the two dots forming the QDM allow one to bring the radiative decay rates on the scale of interdot phonon-assisted relaxation [13]. In this regime, the interband transitions couple to the low energy tails of phonon spectral densities. The carrier-phonon coupling depends sensitively on the geometry of the charge carrier wave functions [19]. Hence, the resulting relaxation dynamics depends on the morphology of the system. In fact, the details of the coupling can in some cases be inferred from the observed phonon-induced carrier dynamics manifested in the optical response [20].

Carrier-phonon interaction in quantum dots (QDs) is governed by the commensurability between the phonon wavelength and the QD geometry: short-wavelength phonons do not couple efficiently, since the deformation or piezoelectric field averages out over the mesoscopic effective volume of the wave function. On the other hand, in a relaxation process, the frequency of the emitted phonon is fixed by energy conservation. The joint effect of this selectivity in frequency and wave vector space leads to suppression of phonon-induced relaxation if the energy separation between the orbital states

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corresponds to wavelengths below the dot size, which typically corresponds to a few meV in self-assembled structures. For anisotropic structures, such as the pancake-shaped QDs to be considered here, the cut-off wavelength depends on the direction of emission, with relatively short-wavelength (few meV energy) phonons emitted almost exclusively along the strongest confinement (growth) axis of the dot.

For a QDM, the situation becomes even more interesting, as relaxation between states primarily localized in different dots is efficient only when one QD is positioned at an antinode of the strain or piezoelectric field generated by the phonon emitted from the other. This leads to oscillations, or resonances, in the QDM relaxation rates as a function of the distance between the QDs. They are manifested particularly clearly for vertically stacked flat QDs with an energy separation in the meV range, which assures an effectively one-dimensional emission along the stacking axis. This prediction has been confirmed by quantitative theoretical studies of phonon-induced relaxation in weakly confined vertically stacked QDMs [21] as well as for both electrons and holes in strained self-assembled structures, where a reliable description must be based on accurate wave function modeling, e.g., using the $\mathbf{k} \cdot \mathbf{p}$ theory [22–24]. Structured phonon relaxation rates resembling the theoretically predicted oscillations have also been observed experimentally [20,25] but the lack of precise modeling at the foundations of the full kinetic description prevented the experimental and theoretical results from being fully reconciled.

Here, we combine experiment and theory to understand signatures of combined carrier-phonon-photon kinetics in which both dissipative processes have comparable rates. We measure the lifetime of the neutral exciton (X^0) in an electrically tunable QDM and observe nonmonotonic behavior as a function of the applied voltage. Our kinetic model incorporates quantitative modeling of carrier states and phonon spectral densities using $\mathbf{k} \cdot \mathbf{p}$ theory based on a precise mapping of the QDM morphology [26]. This allows us to relate this feature to the interplay between phonon-assisted relaxation and radiative recombination in a regime where relaxation is slowed down by tuning the energy separation between the two lowest X^0 eigenenergies in the QDM to the oscillating tail of the phonon spectral density. We restrict our discussion to the standard Fermi golden rule description, which has been successful in accounting for the main features of the relaxation process, although higher-order and non-Markovian corrections may become observable under certain conditions [27]. The observed antiresonances, where the phonon-mediated coupling of orbital and spin states are strongly suppressed, can be used to circumvent phonon-mediated relaxation and decoherence processes [28,29]. This tunability of the lifetime can be leveraged to, e.g., increase the lifetime of energetically unfavorable charge configurations needed for realizing efficient spin-photon interfaces and multidimensional cluster states for quantum communication and measurement based quantum computation [4–7].

The paper is organized as follows: In Sec. II, we describe the sample design, experiments, and results of the voltage dependent time resolved measurements. In Sec. III, we define the theoretical model, which is used to quantitatively interpret the experimental results. Finally, Sec. IV contains the con-

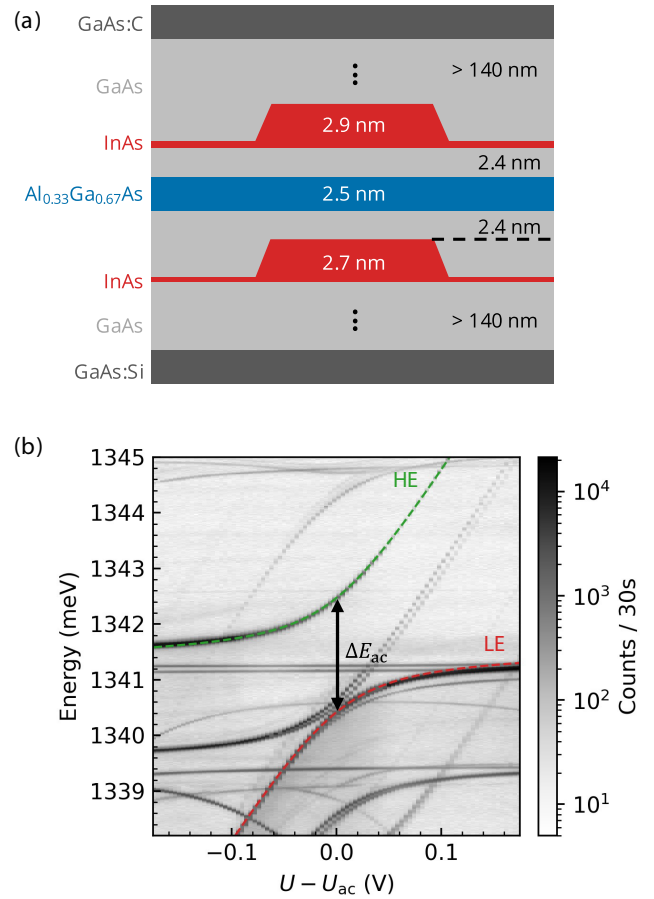


FIG. 1. (a) Schematic structure showing the central part of the QDM sample embedded in a p - i - n diode. (b) Voltage-dependent photoluminescence spectra of a QDM. The neutral exciton eigenenergies (HE in green and LE in red) were fitted by a two-state effective Hamiltonian [see Eq. (A1)]. ΔE_{ac} is the energy splitting at the avoided crossing at $U_{ac} = 0.58$ V.

clusions and compares the experiment with the theory. In the Appendix, we provide details related to the experimental and theoretical parts. We also present the effective model utilized to extract the exciton kinetics.

II. EXPERIMENT

In this section, we describe the experimental methods and present the measurement results.

A. Sample structure and methodology

Figure 1(a) shows the (epitaxial) layer design of the QDM sample, consisting of two vertically stacked InGaAs QDs embedded in a GaAs matrix and grown using the In-flush method to precisely control their absolute height. The interdot separation was controlled during growth to be 10 nm (wetting layer of both dots), resulting in electron tunnel-coupling between the dots forming the molecule [30]. The QDM is embedded in the intrinsic region of a p - i - n diode allowing the application of a gate voltage U to the sample to control the static electric field along the QDM growth axis. The $\text{Al}_{0.33}\text{Ga}_{0.67}\text{As}$

layer between the two dots acts as a tunnel barrier, thereby reducing the tunnel coupling strength. Further information on the growth and fabrication of the sample can be found in Appendix 1 and in previous work in Refs. [2,31].

We optically address the fundamental neutral exciton transitions (X^0) as their energy splitting is tuned to explore signatures of interplay between the phonon-induced and radiative kinetics. In all measurements presented here, we make use of a two-step electrical and optical sequence: First, we efficiently prepare the QDM in a zero-charge (empty QDM) ground state by applying a large negative voltage of -6 V. In a subsequent measurement step, we apply a static gate voltage [3] to tune the electronic orbital states in each of the two QDs relative to each other. The top dot has a larger height, thereby ensuring that the hole of an excited exciton is always located in the top dot. In this way, the spatially direct (electron in top dot) and indirect (electron in bottom dot) exciton energies can be tuned into resonance.

To perform luminescence lifetime measurements, the X^0 is excited using a 5 ps laser pulse tuned to 1362.1 meV to be resonant to the p shell in the upper dot. After a few picoseconds, the system relaxes nonradiatively to the energetically lower s -type exciton states, which enables the detection of the X^0 -shell emission and its energy and lifetime. See Appendix 2 for further details on the experiment and setup.

B. Experimental results

The gate voltage (U) dependent photoluminescence spectra of a QDM excited with an above band-gap laser is presented in Fig. 1(b). The different emission lines belong to different charged states, including the neutral exciton (X^0), negatively charged trion (X^-), and doubly negatively charged trion (X^{2-}). Since each charge state exhibits unique transitions, their spectral features enable their identification. The origin and appearance of these spectral features have been studied intensively in previous work [32–36].

The two energy branches belonging to the neutral exciton X^0 are henceforth referred to as the low energy (LE) branch and the high energy (HE) branch. The weak parabolic voltage dependent shifts for LE and HE correspond to excitonic states having direct character (e and h in same dot) while strongly linearly shifting transitions are spatially indirect excitons (e in lower dot, h in upper dot) that exhibit a large linear Stark shift due to their significantly larger static dipole moment. At $U_{ac} = 0.58$ V, the electron orbitals of both dots are in resonance and electron tunnel coupling results in a pronounced avoided crossing U_{ac} between the orbital states, shown by ΔE_{ac} in Fig. 1(b). Tracing through the avoided crossing corresponds to a transition between the spatially direct and indirect exciton states. This reflects the well-known structure of the tunnel resonance, where the electron envelope function changes its localization from one dot to the other upon moving through the anticrossing [1,2,30,37]. At the point of minimal energy splitting $\Delta E_{ac} = 1.98$ meV, the electron envelope functions hybridize via coherent tunnel coupling to form bonding and antibonding superpositions of the uncoupled orbitals.

The exciton energies are fitted using a coupled two-state effective model including the dc Stark shift for inter-dot excitons [1,30]. The full description can be found in

Appendix 3. The eigenstates of the effective model are shown in Fig. 1(b) by the green and red dashed lines exhibiting good agreement with experiment. In the following, the results of the effective model are used to calculate the radiative decay rates of the HE and LE branches, allowing us to relate the phonon resonances to the energy splitting between coupled exciton states.

To probe the acoustic phonon couplings, time-resolved decay transients of the X^0 states were measured for various gate voltages around the avoided crossing. In this region, the energy difference between HE and LE ranges from ≈ 2 meV to 10 meV, making an acoustic phonon absorption or emission at 1.7 K possible. Figures 2(a) and 2(b) show two representative decay curves: one recorded from the low-energy branch at $U - U_{ac} = 0.32$ V and the other from the high-energy branch at $U - U_{ac} = -0.08$ V. The initial increase in the occupation for both cases comes from nonradiative transitions from the higher orbital states (p shell). Figure 2(a) shows a typical time resolved decay transient recorded from the low-energy branch (black). The relaxation time is obtained by fitting with a monoexponential function convolved with a Gaussian (red). The exciton decay time of the high-energy branch [see Fig. 2(b) in black] exhibits two distinct slopes, corresponding to a fast τ_F^{-1} and a slow τ_S^{-1} relaxation time. Their decay rates are obtained by fitting with a double-exponential decay convolved with a Gaussian function (green). Details on the fitting functions and the convolution with an instrument response function can be found in Appendix 4.

The measured decay times depend not only on the radiative lifetimes of the coupled orbital states but also on other processes that depopulate the HE and LE states. While we will show below that these processes reflect the coupling to acoustic phonons, we first calculate the radiative recombination rates of the HE and LE branches. Therefore, we measure the radiative lifetime of the direct exciton by applying gate voltages far from any resonances and the avoided crossing, e.g., at $U - U_{ac} = 0.62$ V, where the level splitting is approximately 19 meV. Hereby, we suppress transitions mediated by acoustic phonons since the coupling matrix element vanishes for such large phonons as their wavelength is far below any critical dimension reflecting the envelope functions. As a result, the measured decay rate $\tau_D^{-1} = 2.45$ ns $^{-1}$ for the LE branch can be interpreted as the radiative recombination rate of the direct exciton, $\Gamma_D = \tau_D^{-1}$. The recombination rate for the indirect exciton, $\Gamma_I = 0.04$ ns $^{-1}$, was estimated from the electric field dependent Rabi oscillation frequency, as reported in Ref. [2]. Using these determined values Γ_D and Γ_I far away from the avoided crossing, the radiative recombination rate can be calculated at arbitrary voltage in the spirit of the effective model by weighting the recombination rates for the direct and indirect excitons according to the Hopfield coefficients of the coupled system,

$$\Gamma_n(U) = |c_n^{(D)}(U)|^2 \Gamma_D + |c_n^{(I)}(U)|^2 \Gamma_I, \quad (1)$$

where $n \in \{\text{LE}, \text{HE}\}$ labels the exciton energy branches, U is the gate voltage, and $c_n^{(D/I)}(U)$ are coefficients resulting from the diagonalization of the coupled two-state Hamiltonian [Eq. (A1) in Appendix 3]. The results of this model are represented by the solid green (HE) and red (LE) lines in Figs. 2(c)

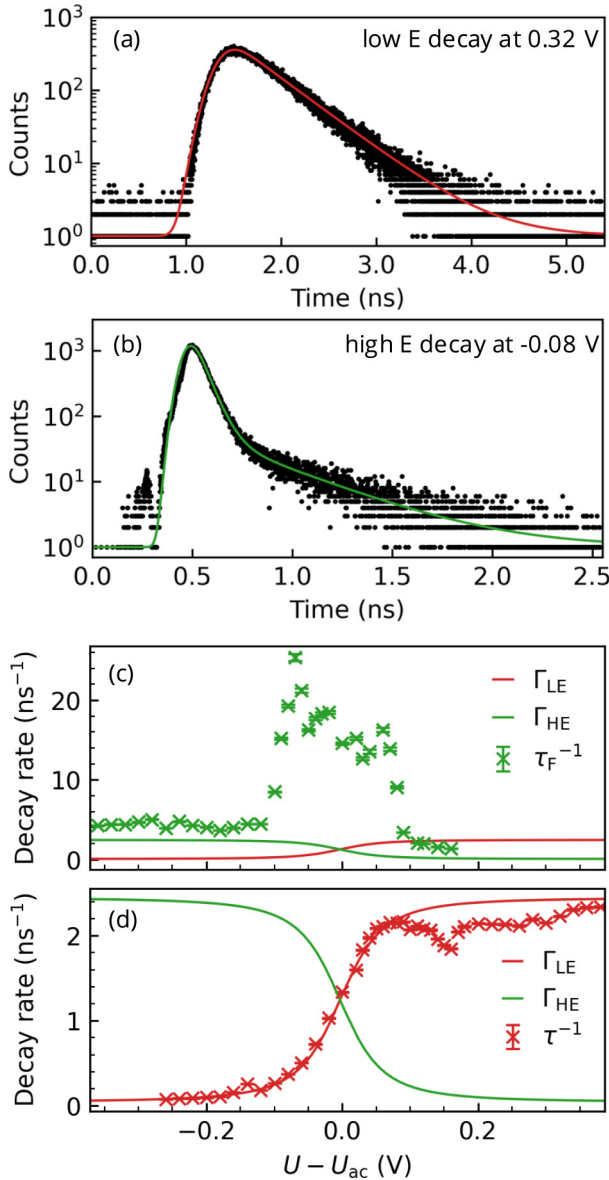


FIG. 2. The time dependence of the emission from the two low-energy neutral exciton energy branches: (a) The exciton decay time from the lower energy branch at 0.32 V from the avoided crossing and the fitting using an exponential function convolved with a Gaussian to account for the instrument response (IR). (b) Results from the higher energy branch at -0.08 V from the avoided crossing and the fitting by a sum of two exponentials convolved with a Gaussian. (c), (d) Gate voltage dependent decay times. The fast decay rates τ_F^{-1} of the higher energy branch are shown with green symbols in (c), the decay rates τ^{-1} of the lower energy branch with red symbols in (d). The red and green solid lines denote the radiative recombination rates Γ_{LE} and Γ_{HE} of LE and HE calculated from Eq. (1), respectively.

and 2(d). For gate voltages far below (above) the avoided crossing U_{ac} , the LE (HE) branch exhibits a strong indirect exciton character, leading to slow radiative recombination. As the voltage increases (decreases), the overlap between the electron and hole wave functions increases, resulting in faster decay. For gate voltages far above (below) the avoided crossing, the LE (HE) state is predominantly a direct exciton

state exhibiting a fast radiative recombination. The crossing of the solid lines marks the hybridization of the electron wavefunction over both dots, as also illustrated by ΔE_{ac} in Fig. 1(b).

The extracted decay rates of the high-energy (green crosses) and low-energy (red crosses) branches are shown in Figs. 2(c) and 2(d) for gate voltages ranging from -0.36 V to 0.38 V around the avoided crossing U_{ac} . As described earlier, τ^{-1} for the low-energy branch is obtained by fitting a monoexponential decay. The measured values of τ^{-1} closely follow the dependence predicted by the effective model for radiative decay, $\Gamma_{LE}(U)$. However, at approximately 0.14 V, a pronounced dip appears, corresponding to an HE-LE energy splitting of 4.8 meV. The origin of this phonon-related feature is discussed in detail in Sec. III C.

The high-energy branch is modeled using a double-exponential decay, incorporating both τ_F^{-1} (fast decay) and τ_S^{-1} (slow decay) components. The lifetime τ_F (fast decay) corresponds to the radiative decay of the HE state, further enhanced by phonon-assisted relaxation to the LE state. As shown in Fig. 2(c), the measured values of τ_F^{-1} deviate from the predicted radiative decay rates, $\Gamma_{HE}(U)$. The pronounced peaks and phonon-related features at -0.07 V and 0.08 V correspond to an HE-LE energy splitting of 2.75 meV and will be discussed in detail in Sec. III C. The second lifetime, τ_S (slow decay), is associated with transitions from the LE state to the HE state via acoustic phonon absorption. The impact of this process is proportional to the occupation of the LE state, which itself depends on the radiative recombination rate $\Gamma_{LE}(U)$. Consequently, the τ_S^{-1} rate observed in the HE branch provides insights into exciton dynamics within the LE branch. However, at 1.7 K, acoustic phonon absorption is very unlikely. Therefore, the slower decay is only visible for a few voltages. Since the optical count rates for this slow decay are very low at 1.7 K [see Fig. 2(b)], a precise extraction is not possible. To validate this physical picture, we conducted lifetime measurements on a different QDM sample at a higher temperature of 10 K, where acoustic phonon absorption is more pronounced due to the underlying Bose-Einstein statistics. Figure 7 in Appendix 5 presents this additional measurement, showing the slow decay rate for all measured HE decays.

In the following, we provide a detailed analysis of the mismatch between the radiative decay rates predicted by the effective model and those measured experimentally, attributing the discrepancy to phonon resonances that occur when the q vector of the involved phonon matches a characteristic length scale in the QDM.

III. THEORY

In this section, we briefly describe the theoretical models utilized in the numerical simulations. The calculation details are presented in Appendix 6. We then continue to discuss the relation between the theoretical and experimental results.

A. The theoretical framework

We represent the shape of the QDs as truncated Gaussians [38]. The model adopted in the calculations is shown

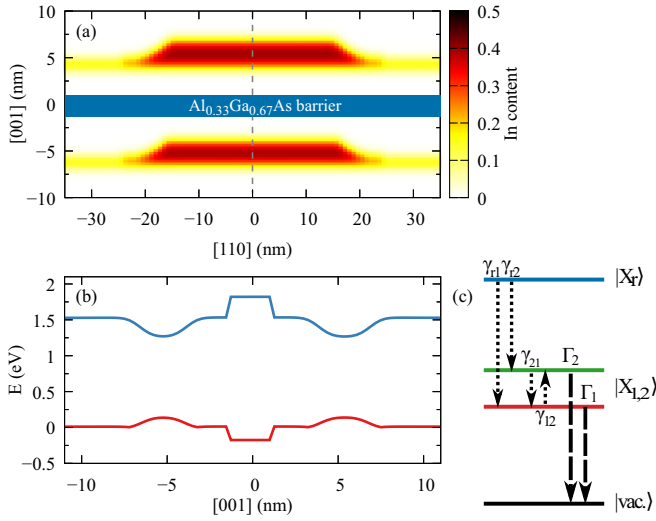


FIG. 3. (a) The model of In distribution in the structure and the $\text{Al}_{0.33}\text{Ga}_{0.67}\text{As}$ barrier. (b) The conduction band (blue) and heavy-hole band edges (red) along the axis crossing the QDs centers [as marked in (a) by the dashed line]. (c) Schematic structure of states and processes driving the system dynamics.

in Fig. 3(a) and inspired by scanning transmission electron microscopy images presented in Ref. [39].

The lattice mismatch between the QDs material (InGaAs) and the GaAs results in strain in the system. We obtain the strain distribution within the continuous elasticity approach [40] by minimization of the elastic energy. As the lattice constants of AlAs and GaAs are very similar, the AlGaAs barrier is neglected at this stage of simulations. We calculated the piezoelectric potential by taking the strain-induced polarization up to second order in the strain tensor elements [41] using parameters from Ref. [42].

The electron and hole single-particle states in the QDM are calculated using the eight-band $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian [43,44] in the envelope function approximation. The details of the implementation are given in the Appendix of Ref. [26]. We account for the $\text{Al}_{0.33}\text{Ga}_{0.67}\text{As}$ barrier in the $\mathbf{k} \cdot \mathbf{p}$ calculation by altering the band gap and valence band offset. To visualize the effects of the barrier and strain, we compute the Γ -point band edges by diagonalizing the eight-band Bir-Pikus Hamiltonian for virtual bulk systems with compositions and strain equal to those in the actual system along a line passing through the centers of the QDs [see Fig. 3(b)]. The single-particle wave functions form a basis for further calculations of the neutral exciton states. The latter has been done within the configuration-interaction (CI) method. At this stage, we also include the electric field (the Stark effect) into the model, as described in Appendix 6.

To obtain the radiative relaxation times of the exciton states, we calculate their oscillator strength [45,46]. This is performed using the interband momentum matrix elements, determined within the Hellman-Feynman theorem [47–49] with the eight-band $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian.

The phonon-assisted transition rates between the exciton states were calculated within Fermi's golden rule. Here, we take into account the coupling via deformation poten-

tial and piezoelectric field [50,51]. The details are given in Appendix 6.

Even though both the QD morphology and material parameters suffer from some uncertainty, we believe we can achieve reasonable final accuracy of the results by working in a feedback loop in which the model is confronted with the experimental data at least at three levels: QDM geometry input, spectroscopic data, and kinetic rates.

B. The system dynamics

The calculated quantities constitute a starting point for studying the kinetics of the system. A schematic structure of the relevant states and processes driving the system is shown in Fig. 3(c). In the following, for the sake of readability of the formulas, we employ a simplified notation in which subscripts 1 and 2 are used in place of LE and HE, respectively. Accordingly, the states are written as $|X_{1,2}\rangle$, denoting the LE and HE states. $|X_r\rangle$ is the reservoir of the higher states of the system (the p shell in the experiment). The states $|X_{1,2}\rangle$ are fed from the reservoir via a very fast decay with rates γ_{r1} and γ_{r2} . This provides an effective description of the states excited via the p - p excitons. The $|X_{1,2}\rangle$ states decay to the ground (vacuum) state via radiative decay with rates Γ_1 and Γ_2 . In addition, phonon emission and absorption processes induce transitions between these two states with rates γ_{12} and γ_{21} . This kinetics is described by the rate equations

$$\frac{dN_0}{dt} = \Gamma_1 N_1 + \Gamma_2 N_2, \quad (2a)$$

$$\frac{dN_1}{dt} = \gamma_{r1} N_r + \gamma_{21} N_2 - (\gamma_{12} + \Gamma_1) N_1, \quad (2b)$$

$$\frac{dN_2}{dt} = \gamma_{r2} N_r + \gamma_{12} N_1 - (\gamma_{21} + \Gamma_2) N_2, \quad (2c)$$

$$\frac{dN_r}{dt} = -(\gamma_{r1} + \gamma_{r2}) N_r, \quad (2d)$$

where N_0 is the occupation of the vacuum state; N_1 , N_2 are the occupations of the two lowest exciton states (neglecting the fine structure), and N_r accounts for the occupation of the state representing the reservoir. The set of differential equations is solved numerically, and the resulting occupations are convolved with a Gaussian instrumental response function, as described in Appendix 6.

C. Results and comparison to experiment

The results of the theoretical model and the experimental values for the exciton energy splitting are presented in Fig. 4(a). The theoretical value for the energy splitting at the avoided crossing ($F_{ac} = 18.3$ kV/cm) is $\Delta E_{res} = 1.97$ meV. We find overall very good agreement with the experimental data (marked by green points) with the avoided crossing at $U_{ac} = 0.58$ V and an energy splitting of $\Delta E_{ac} = 1.98$ meV. Further, the relative voltage axis can be converted to an relative electric field axis via

$$F - F_{ac} = -\frac{U - U_{ac}}{D}, \quad (3)$$

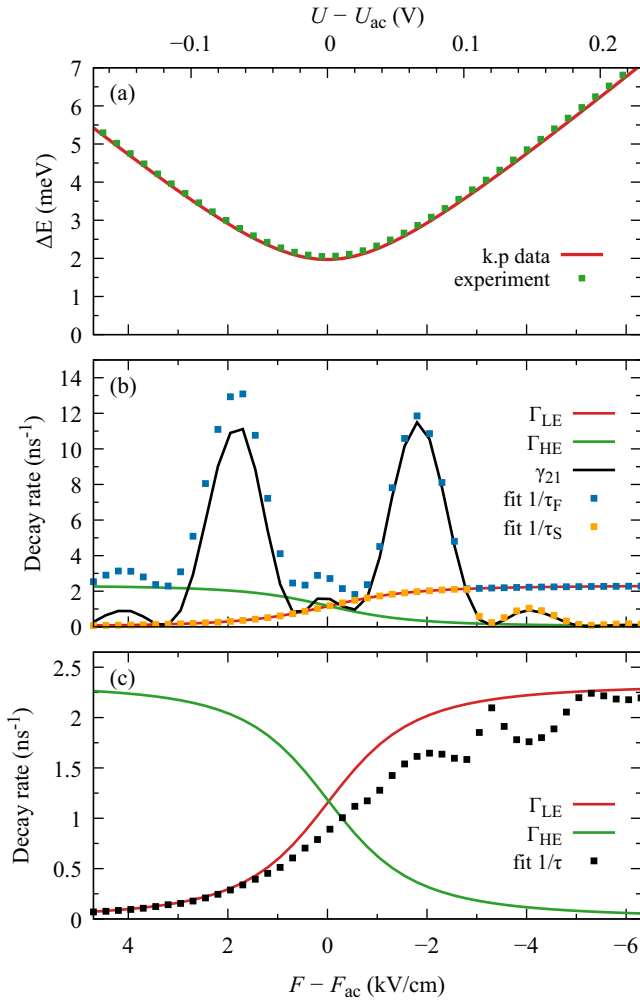


FIG. 4. (a) The theoretical (experimental) results for the difference between the two-lowest exciton energy branches as a function of the axial electric field (gate voltage). (b) The electric-field dependence of the relaxation times (τ_F^{-1} and τ_S^{-1}) fitted to the numerical $k \cdot p$ results via the convoluted biexponential decay. The green and red solid lines denote the radiative recombination rates calculated directly from the exciton states, and the black solid line denotes the phonon assisted relaxation rate from HE to LE. (c) The decay rates τ^{-1} were obtained via the convoluted monoexponential decay. The electric field axis was inverted to facilitate the comparison to experimental data, which are presented as a function of the gate voltage.

with U_{ac} and F_{ac} as above, and $D = 365$ nm being the distance between the two gates (n and p regions). For comparison, both axes are centered at the avoided crossing.

We calculated the radiative recombination rates Γ_n and the rates of transitions mediated by the acoustic phonons γ_{nm} . The radiative recombination rates Γ_{LE} and Γ_{HE} for the LE and HE branches obtained from the calculated exciton states [Eq. (A7)] are shown in Figs. 4(b) and 4(c) by the solid red and green lines. As observed from the values far from the avoided crossing, the radiative recombination rate of the direct exciton is $\Gamma_D = 2.33$ ns⁻¹, which is close to the measured value of 2.45 ± 0.008 ns⁻¹. We calculated the phonon-assisted relax-

ation rate from the HE to the LE exciton branch (γ_{21}) as a function of the electric field. This dependence shows an oscillatory behavior, which is linked to the underlying phonon spectral density [52]. The calculated phonon-assisted relaxation rate γ_{21} reaches values of up to approximately 11 ns⁻¹ at $F - F_{ac} = 1.8$ kV/cm and $F - F_{ac} = -1.8$ kV/cm, which is significantly faster than the radiative decay. These resonances correspond to an energy splitting of the HE and LE branches of $\Delta E \approx 2.8$ meV. However, for electric fields far from the avoided crossing, the overlap between the electron wave functions decreases, causing the γ_{21} rate to vanish. The positions of the second maxima in γ_{21} are $F - F_{ac} = 4.1$ kV/cm and $F - F_{ac} = -4.1$ kV/cm, both corresponding to an energy splitting of $\Delta E \approx 4.8$ meV.

Due to interference, the phonon-assisted transition rates are expected to have maxima if $D_{QD} = (n + 1/2)\lambda_{ph}$, with λ_{ph} being the phonon wavelength and $D_{QD} = 10$ nm the distance between the QDs. Accordingly, the acoustic phonon energy matches $\hbar\omega = \hbar(2n + 1)\pi c_1/D_{QD}$ [52], where c_1 is the longitudinal speed of sound. Therefore, the maxima are expected for the energy splittings: 1.1 meV ($n = 0$), 3.2 meV ($n = 1$), and 5.3 meV ($n = 2$). While the first energy is below the minimum energy splitting between the HE and LE energy branches, the two others are comparable to $\Delta E = 2.8$ meV and $\Delta E = 4.8$ meV [see Fig. 4(b)]. The corresponding acoustic phonon wavelengths can be obtained via $\lambda_{ph} = \hbar c_1 q = 2\pi \hbar c_1 / \Delta E$. This gives $\lambda_{ph} = 7.6$ nm for $\Delta E = 2.8$ meV and $\lambda_{ph} = 4.4$ nm for $\Delta E = 4.8$ meV, which lies in the characteristic scale of the system.

To compare the measured decay rates shown in Fig. 2 with our model, we solve the rate equations (2a)–(2d) for the four-level system $[N_i(t)]$ using the previously calculated values of Γ_n and γ_{nm} . The values of γ_{r1} , γ_{r2} were taken from the effective model [see Eq. (A9)]. We then apply the convolution [Eq. (A10)] with the instrument response function $f(t)$ [see Eq. (A11)], using the same parameter values as in the experimental fitting, obtaining $\tilde{N}_i(t)$. To ensure a consistent comparison between the exciton dynamics predicted by the full theoretical model and the experimental results, the same fitting scheme must be applied in both cases.

For the HE branch, we fitted $\tilde{N}_i(t)$ using a sum of two exponentials convolved with a Gaussian [see Eq. (A3) in Appendix 4] with the faster (τ_F^{-1}) and with the slower (τ_S^{-1}) decay. The results are shown in Fig. 4(b). The obtained fast decay rate τ_F^{-1} follows the behavior of the phonon-assisted relaxation rate γ_{21} , as expected. The two maxima at $F - F_{ac} = 1.8$ kV/cm and $F - F_{ac} = -1.8$ kV/cm, corresponding to $\Delta E \approx 2.8$ meV, align very well with the two peaks observed at -0.07 V and 0.06 V in the experimental data shown in Fig. 2(c). This agreement provides clear evidence of experimentally observed phonon resonances. However, in the experiment, the decay rate between the two peaks is not reduced as significantly as predicted by the model. This can be related to the fact that the phonon spectral density crucially depends on the detailed shape of the wave function (hence the specific QD morphology) [53], and the model approximates the exact material distribution and the geometries of both QDs forming the molecule. We note, that for the HE branch the radiative and phonon-induced decays behave additively, i.e., $\tau_F^{-1} \approx \Gamma_{HE} + \gamma_{21}$ and $\tau_S^{-1} \approx$

$\Gamma_{\text{LE}} + \gamma_{12}$ for $F - F_{\text{ac}}$ larger than -2.8 kV/cm (and opposite otherwise).

For the LE branch, we fit $\tilde{N}_i(t)$ with a single exponentially modified Gaussian [see Eq. (A2) in Appendix 4]. As shown in Fig. 4(c), the obtained decay rate τ^{-1} is generally consistent with the value of the radiative lifetime Γ_{LE} as expected. However, similarly to the experimental results presented in Fig. 2(d), some deviations appear on the right of the avoided crossing. In Fig. 4(c), two dips are shown. These features appear over $F - F_{\text{ac}}$ range of -2.1 kV/cm to -5.0 kV/cm, which corresponds to exciton energy splittings ΔE between 3.0 meV and 5.7 meV. This is consistent with the position of the dip (antiresonance) observed in the experimental data ($\Delta E = 4.8$ meV at 0.14 V), clearly demonstrating the experimental observation of phonon-related features. It should be noted that in the theoretical model, phonons are treated assuming the GaAs bulk dispersion. However, the actual dispersion in a strained heterostructure may be different, which could lead to a case where only one dip is visible in the experiment. In contrast to the HE branch, the additive behavior of different decay channels breaks down here, as the LE exciton state is repopulated with the rate comparable to Γ_{LE} , which results in joint decay rates that are nontrivial combinations of the original phonon relaxation and radiative recombination rates.

IV. CONCLUSIONS

In our joint experimental-theoretical work, we present an investigation of phonon-mediated processes in an electrically tunable self-assembled QDM. Through voltage-dependent measurements, we directly measure the orbital phonon relaxation rates between the energy states of the neutral exciton and benchmark our findings against microscopic $\mathbf{k} \cdot \mathbf{p}$ calculations based on an accurate mapping of the QDM morphology. Our results reveal phonon-mediated relaxation rates that exhibit pronounced resonances and antiresonances, with rates ranging from tens of μs^{-1} up to tens of ns^{-1} . Comparison with a kinetic model reveals the voltage (energy) dependent phonon coupling strength and fully explains the interplay between phonon-assisted relaxation and radiative recombination. The resonances (antiresonances) observed in the relaxation rates as a function of applied voltage are attributed to enhancement (suppression) of phonon coupling for specific energies. These features enable further tunability of the lifetime which can be leveraged to increase the lifetime of energetically unfavorable charge configurations needed for realizing efficient spin-photon interfaces and multidimensional cluster states [4–7]. Moreover, suppression of orbital-phonon coupling is likely to be associated with inhibited phonon-mediated decoherence of the quantum states and therefore an essential step toward future quantum information applications [8].

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX

1. Sample

The QDM investigated here is fabricated using solid-source molecular beam epitaxy, comprising two vertically stacked indium arsenide (InAs) QDs embedded within a gallium arsenide (GaAs) matrix. The heights of the top and bottom QDs were precisely controlled at 2.9 nm and 2.7 nm, respectively, through the In-flush technique during the growth process [55]. This specific height configuration enables electric tuning of the orbital states in the conduction band when operating the diode in reverse bias [56]. The top dot has a larger height to ensure that the hole is always located in this dot. The separation between the wetting layers of both dots is 10 nm. An aluminum gallium arsenide ($\text{Al}_{0.33}\text{Ga}_{0.67}\text{As}$) barrier with a thickness of 2.5 nm is placed between the QDs, thereby influencing the coupling strength. Additionally, a 50 -nm-thick $\text{Al}_{0.33}\text{Ga}_{0.67}\text{As}$ tunnel barrier was deposited 5 nm below the QDM to extend electron tunneling times out of the QDM.

The QDM is integrated into a p - i - n diode structure, with the doped regions serving as electrical contacts for gating purposes. The diode contacts are situated more than 150 nm away from the QDM to mitigate uncontrolled charge tunneling into the QDM. Furthermore, a $\text{Al}(\text{Ga})\text{As}/\text{GaAs}$ distributed Bragg reflector was incorporated below the diode, while a circular Bragg grating was deterministically positioned above a single preselected QDM using *in situ* electron beam lithography to maximize photon in-coupling and out-coupling efficiencies [57].

2. Experimental details

a. Experimental setup

All measurements discussed in the main text were performed at a temperature of 1.7 K within an Attodry2100 dry magnet system, employing a pulsed fs laser (Coherent Mira

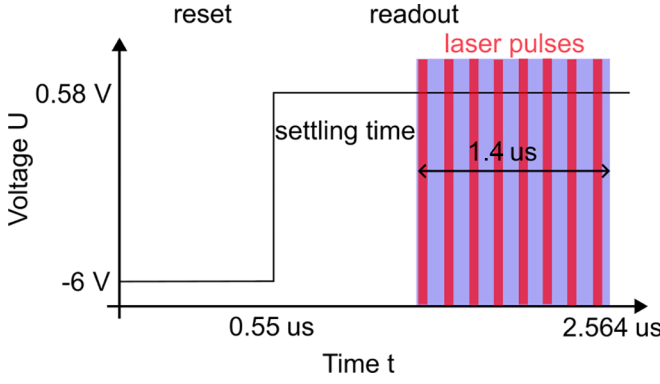


FIG. 5. In the reset phase, a large negative voltage of -6 V is applied for 550 ns to empty the QDM. In the read phase, a static gate voltage is applied to tune the electronic orbital states in each of the two QDs relative to each other (e.g., 0.58 V). After a settling time of approximately 600 ns, the pulsed laser is on and excites the crystal ground state to the p orbital of the neutral exciton (p -shell excitation) for 1.4 μ s.

900f) for excitation of the QDM. The measurements were performed with a repetition rate of 390 kHz corresponding to approximately 2.564 μ s. We make use of a two-phase electrical and optical sequence which is shown in Fig. 5 and described in the following: First, we efficiently prepare the QDM in a zero-charge (empty QDM) ground state by applying a large negative voltage of -6 V for 550 ns (reset phase). In the second phase (read phase), we apply a static gate voltage to tune the electronic orbital states in each of the two QDs relative to each other, e.g., 0.58 V. After a settling time of approximately 600 ns, a 5 ps laser pulse is applied to excite the crystal ground state ($|vac\rangle$) to the p orbital of the neutral exciton (p -shell excitation) for 1.4 μ s. The phonon-mediated relaxation process between the p shell and the s -shell occurs on timescales of a few picoseconds [58–60], while the excitonic lifetime ranges from several hundreds of picoseconds to a few nanoseconds [13,61]. Therefore, the direct radiative emission from the p shell is weak compared to the s -shell emission we are measuring here.

The occupations of the HE and LE states of the neutral exciton are monitored by detecting the photons emitted by the driven two-level system. The photons are detected using a time tagger and single photon detectors. Hereby, we use single-photon avalanche diodes (SPADs) and superconducting nanowire single-photon detectors (SNSPDs). The SPAD has a timing jitter of about 150 ps. Since the measured lifetimes are reaching the limit of our SPAD time resolution, the faster decay rates are measured with a SNSPD with a jitter of about 15 ps. For the low E decay times below -0.12 V, a pulse picker acousto-optical modulator (AOM) was used to excite the QDM with only every second or fourth laser pulse due to the long lifetime of the indirect exciton states. All other measurements were performed without a pulse picker AOM.

b. RC time constants

We measured the RC time constant of the diode to be $RC \approx 100$ ps with $R = 1$ k Ω and $C \approx 100$ fF by using a cryogenic probe station. The resistance R of the diode is

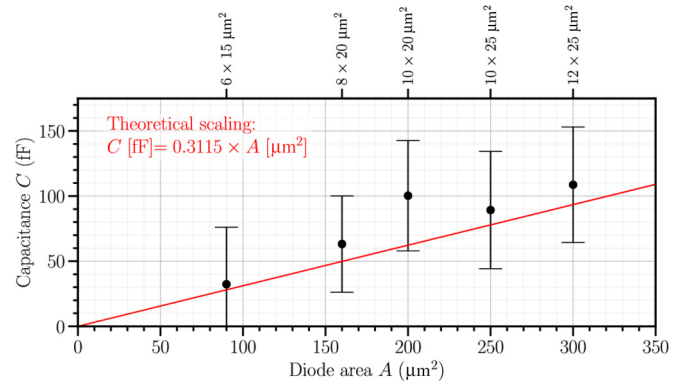


FIG. 6. The measured capacitance (black data points) and calculated values (red line) for different diode areas A . The theoretical scaling relationship is $C[\text{fF}] = 0.3115A$ (μm). The diode measured within this paper has an active area of $10 \times 25 \mu\text{m}^2$ and therefore a capacitance of $C \approx 100$ fF.

an intrinsic property resulting from both the layer design as well as the contacting scheme, and it depends on the diode's operation regime. Since we operate the diode in reverse bias, high resistance is expected. The capacitance C depends on the diodes active area A , the distance between the p and n layers, and the dielectric constants of the materials. Among these parameters, only the active diode area A can be manipulated after growth. Therefore, achieving low capacitance can only be accomplished through a small diode area. Figure 6 shows the measured capacitance (black data points) and calculated values (red line) for different diode areas, with the theoretical scaling relationship $C[\text{fF}] = 0.3115A$ (μm). The diode measured within this paper has an active area of $10 \times 25 \mu\text{m}^2$.

In our experimental setup, the diode is contacted via coaxial cables to a dc voltage source. These cables add a parasitic capacitance and increase the RC time constant. However, at our repetition rate of 390 kHz and AWG switching times of <20 ns, we are not limited by the RC time constant of the experiment. We can confirm this by analyzing the linewidth of the indirect exciton (<50 μeV), which shows no broadening due to excessively fast electrical switching of the diode. Additionally, we ensure complete settling of any transient effects by implementing a 0.2 μ s delay (here to be sure we even used a 0.6 μ s delay as shown in Fig. 5) before turning the laser on in the read phase.

3. Coupled two-state Hamiltonian

The neutral exciton in our system can be described by the following effective two-state Hamiltonian:

$$H_{\text{eff}} = \begin{pmatrix} \varepsilon_I + \alpha eU & V_t \\ V_t & \varepsilon_D \end{pmatrix}, \quad (\text{A1})$$

where ε_I , ε_D are the energies of the indirect and direct exciton states when the tunnel coupling and external electric field are neglected. The dependence on the gate voltage (U) enters via the term αeU . The tunnel coupling is represented by the parameter V_t , which is half of the energy splitting at the resonance $V_t = \Delta E_{ac}/2$. By fitting this to the voltage-dependent photoluminescence spectra for the consid-

ered sample in Fig. 1(b), we obtain the following values: $\varepsilon_D = 1324.1$ meV, $\varepsilon_I = 1341.4$ meV, $\alpha = 3.02 \times 10^{-2}$, and $V_I = 0.992$ meV. Our avoided crossing lies at $U_{ac} = 0.58$ V with an energy splitting of $\Delta E_{ac} = 1.98$ meV.

4. Fit functions of lifetime measurements

a. Exponential convolved with a Gaussian

The lifetime measurements with a monoexponential decay are fitted with an exponential function convolved with a Gaussian (exponentially modified Gaussian) supplemented by a constant C accounting for the background

$$\mathcal{G}_1(t) = \frac{A}{2} e^{\gamma(\mu-t+\gamma\sigma^2/2)} \operatorname{erfc}\left(\frac{\mu + \gamma\sigma^2 - t}{\sqrt{2}\sigma}\right) + C, \quad (\text{A2})$$

where μ is the position of the Gaussian component and σ its standard deviation. $\gamma = \tau^{-1}$ defines the exponential decay, A is a scaling constant, and $\operatorname{erfc}()$ is the complementary error function. The exponential decay arises from an ideal Lorentzian line width of the QD emission, while the Gaussian component is caused by an instrument response function [62]. We obtain $\sigma = 0.16$ ns⁻¹ for the measurements taken with the SPAD and $\sigma = 0.04$ ns⁻¹ for the ones taken with the SNSPDs. μ is the time delay until the first counts are detected and is different for the measurements done with the SPAD and SNSPD as well as the ones with and without pulse picker AOM.

b. Sum of two exponentials convolved with a Gaussian

The lifetime measurements with two exponential components in the decay are fitted by a sum of two exponential functions convolved with a single Gaussian

$$\mathcal{G}_h(t) = \frac{A}{2} e^{\gamma_1(\mu-t+\gamma_1\sigma^2/2)} \operatorname{erfc}\left(\frac{\mu + \gamma_1\sigma^2 - t}{\sqrt{2}\sigma}\right) + \frac{B}{2} e^{\gamma_2(\mu-t+\gamma_2\sigma^2/2)} \operatorname{erfc}\left(\frac{\mu + \gamma_2\sigma^2 - t}{\sqrt{2}\sigma}\right) + C, \quad (\text{A3})$$

where A, B represent the scaling factors for the two decays with the $\gamma_1 = \tau_1^{-1}$ (fast decay) and $\gamma_2 = \tau_2^{-1}$ (slow decay) rates, respectively. All other parameters are defined as above in the monoexponential case.

5. Decay rates at 10 K

The decay rates of the neutral exciton presented in Fig. 2 were measured at 1.7 K. A different QDM with the same layer structure was measured at 10 K exhibiting the same behavior in the voltage dependent decay rates. Due to the higher temperature, upscattering is much more likely, leading to a double exponential decay in the high energy emission with a stronger pronounced slow decay. Figure 7 shows the results.

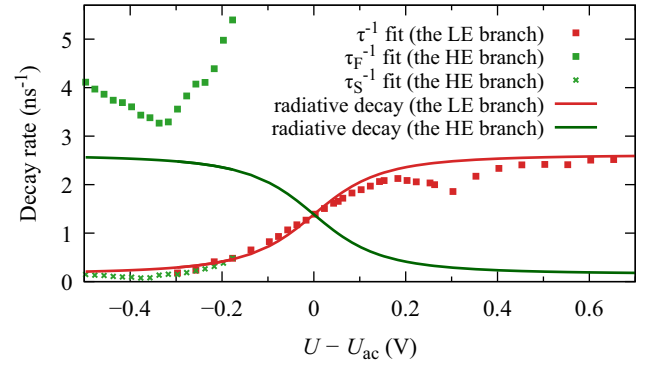


FIG. 7. Voltage dependent lifetime measurements at a temperature of 10 K. The fast and slow decay rates of the higher energy branch are shown in green, the decay rates of the lower energy branch in red. The solid lines denote the radiative recombination rates calculated from Eq. (1).

6. Calculation details

a. QD model and strain distribution

The QDs are modeled as truncated Gaussians [38], where their top surfaces are given by the formula

$$S(x, y) = \min\left(w \exp\left\{-\frac{(x^2 + y^2)}{l^2}\right\}, h\right), \quad (\text{A4})$$

where w is related to the slope steepness, h is the maximal height, and the lateral extension is related to $2l$. For both QDs, we use $l = 20a$ (where a is the lattice constant) and $w = 25a$. The heights are $h_u = 4a$ and $h_l = 3.5a$ (counting from the top of the wetting layer) for the upper and lower QDs, respectively. The thickness of the wetting layer is a . All the geometric parameters are subjected to the discretization scheme with a mesh size of $a = 0.5653$ nm (the barrier lattice constant) for the in-plane directions and $a/2$ for the growth direction. The material composition in the QDs and the wetting layers is In_{0.4}Ga_{0.6}As. To take into account the material intermixing, the InGaAs material distribution is processed by the Gaussian blur with a standard deviation of 0.6 nm. The interdot distance is (counted from the bottom of wetting layer to the bottom of wetting layer) is $17.5a$. Finally, we introduce a $4.5a$ thick Al_{0.33}Ga_{0.67}As barrier between the dots.

b. Configuration-interaction model

The neutral exciton states are calculated using the CI method within the basis of 24 lowest electron and 24 lowest hole states. The implementation description is provided in the Supplemental Material of Ref. [1]. We take into account the Stark effect due to the axial electric field F via a band-diagonal term $V_F(\mathbf{r}) = |e|F(z - z_0)$, where z_0 is the center of the computational box. We calculate the matrix elements of $V_F(\mathbf{r})$ in the basis of single-particle states and incorporate them into the many-body CI Hamiltonian [63]:

$$|X_n\rangle = \sum_{i,j} c_{ij}^{(n)} a_i^\dagger h_j^\dagger |\text{vac}\rangle, \quad (\text{A5})$$

where $|\text{vac}\rangle$ is the vacuum state, and $a_i^\dagger$, $h_j^\dagger$ are the creation operators for the electron and hole, respectively. One should note that exciton energy levels form multiplets containing four states split via the exchange interaction [64]. We here consider one bright configuration per multiplet.

c. Radiative lifetime

The oscillator strength describing the exciton recombination can be written as [45,46]

$$f_n = \frac{2}{m_0 E_n^{(X)}} \sum_{m=x,y,z} \left| \sum_{i,j} c_{ji}^{(n)} \langle \Psi_i^{(v)} | p_m | \Psi_j^{(e)} \rangle \right|^2,$$

where $E_n^{(X)}$ is the energy corresponding to the $|X_n\rangle$ state, $\Psi_i^{(v/e)}$ are the single-particle states from the valence/conduction band. The matrix elements of the momentum operator $(p_m)_{ij} \equiv \langle \Psi_i^{(v)} | p_m | \Psi_j^{(e)} \rangle$ are calculated using the Hellman-Feynman theorem [47–49],

$$(p_m)_{ij} \approx \frac{m_0}{\hbar} \langle \Psi_i^{(v)} | \frac{\partial H_{k,p}}{\partial k_m} | \Psi_j^{(e)} \rangle, \quad (\text{A6})$$

where $H_{k,p}$ is the eight-band $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian. The radiative recombination rate for the state $|X_n\rangle$ can be calculated from [46,65]

$$\Gamma_n = f_n (E_n^{(X)})^2 \frac{n_r e^2}{6\pi \epsilon_0 \hbar^2 m_0 c^3}, \quad (\text{A7})$$

where n_r is the refractive index, ϵ_0 is the vacuum permittivity, and c is the speed of light. For GaAs, we use $n_r = 3.347$ [66].

d. The exciton-phonon interaction

The interactions between the carriers and acoustic phonons are described by the Hamiltonian H_{int} , which can be expressed by terms involving individual phonon modes [50]

$$H_{\text{int}} = \sum_{\mathbf{q}, \lambda} \mathcal{V}(\mathbf{q}, \lambda) e^{i\mathbf{q}\mathbf{r}},$$

where λ are phonon branches (one longitudinal and two transversal) and $\mathbf{q}$ are the phonon wave vectors. In $\mathcal{V}(\mathbf{q}, \lambda)$, we take into account the contributions stemming from the deformation-potential coupling as well as the piezoelectric-field coupling. The complete description of the model can be found, e.g., in Ref. [50]. To calculate the phonon-assisted transitions between the exciton states $|X_n\rangle \rightarrow |X_m\rangle$, we utilize Fermi's golden rule

$$\gamma_{nm} = \frac{2\pi}{\hbar^2} |n_B(\omega_{mn}) + 1| \sum_{\mathbf{q}, \lambda} |F_{nm,\lambda}^{(X)}(\mathbf{q})|^2 \times [\delta(\omega_{mn} - c_\lambda q) + \delta(\omega_{mn} + c_\lambda q)], \quad (\text{A8})$$

where $n_B(\omega)$ is the Bose-Einstein distribution, $\omega_{ij} = \Delta E_{ij}^{(X)}/\hbar = (E_j^{(X)} - E_i^{(X)})/\hbar$, and c_λ is the phonon-branch-dependent speed of sound. The exciton form factor $F_{nm,\lambda}^{(X)}(\mathbf{q})$ is defined in terms of the electron $F_{nm,\lambda}^{(e)}(\mathbf{q})$ and hole $F_{nm,\lambda}^{(h)}(\mathbf{q})$

form factors [67]

$$F_{nm,\lambda}^{(X)}(\mathbf{q}) = \sum_{i,j,i',j'} c_{ij}^{(n)*} c_{i'j'}^{(m)} (F_{ii',\lambda}^{(e)}(\mathbf{q}) \delta_{jj'} - F_{jj',\lambda}^{(h)}(\mathbf{q}) \delta_{ii'}),$$

where

$$F_{ij,\lambda}^{(e/h)}(\mathbf{q}) = \langle \Psi_i^{(e/h)} | \mathcal{V}(\mathbf{q}, \lambda) e^{i\mathbf{q}\mathbf{r}} | \Psi_j^{(e/h)} \rangle.$$

e. Material parameters

We provide the values of material parameters in the Supplemental Material [68]. The elastic constants comes from Vurgaftman *et al.* [69] while the first- and second-order piezoelectric coefficients are taken from Ref. [42]. In the case of the $\mathbf{k} \cdot \mathbf{p}$ simulations, we take most of the parameters (the band gaps, the Luttinger parameters, the deformation potentials) from Ref. [69] and some (e.g., the electron effective mass) from Ref. [43]. To ensure the eight-band $\mathbf{k} \cdot \mathbf{p}$ model remains elliptic, we use the rescaled Kane energy value [70]

$$E_P = \left(\frac{m_0}{m_e^*} - 1 \right) \frac{E_g(E_g + \Delta_0)}{E_g + 2\Delta_0/3},$$

where m_e^* is the electron effective mass, E_g is the band gap, and Δ_0 is the spin-orbit coupling splitting parameter. To represent the AlGaAs barrier, we took the valence band offset $\text{VBO}(\text{AlAs}) = -0.57$ eV and the conduction band offset $\text{CBO}(\text{AlAs}) = 0.87$ eV [71] (with respect to the unstrained GaAs). The GaAs dielectric constant is $\epsilon_r = 12.4$ [72].

In the case of calculations of phonon-induced transitions, we use the GaAs bulk parameters: the piezoelectric coupling is taken in the linear order, hence we take $e_{14} = -0.16$ C/m² [73]. We took the longitudinal and transverse speed of sound $c_l = 5150$ m/s and $c_t = 2800$ m/s, and the crystal density is $\rho = 5350$ kg/m³ [74].

f. The rate equations

The relaxation rates from the reservoir $\gamma_{r,1}$, $\gamma_{r,2}$ that enter the rate equations (dN_i/dt) are calculated from

$$\gamma_{rn}(U) = |c_n^{(D)}(U)|^2 \gamma_r^{(D)} + |c_n^{(I)}(U)|^2 \gamma_r^{(I)}, \quad (\text{A9})$$

where $c_n^{(D)}(U)$, $c_n^{(I)}(U)$ are the effective-model coefficients as in Eq. (1), and $\gamma_r^{(D)}$ ($\gamma_r^{(I)}$) is the intradot (interdot) transition rate between the p - and the s -type states. We took $\gamma_r^{(D)} = 160$ ns⁻¹ and $\gamma_r^{(I)} = 20$ ns⁻¹, owing to the fact that intradot relaxation is much faster than phonon-assisted tunneling.

We numerically solve the set of differential equations using Scipy [75] library (odeint) to obtain the time evolution of the occupations $N_i(t)$. For comparison with the experimental data, the results are convolved with the instrument response function $f(t)$ [62],

$$\tilde{N}_i(t) = \int_{-\infty}^{\infty} f(\tau) \Theta(t - \tau) N_i(t - \tau) d\tau, \quad (\text{A10})$$

where $\Theta(t)$ is the Heaviside step function and

$$f(t) = \frac{1}{\sigma \sqrt{2\pi}} \exp\left(-\frac{(t - \mu)^2}{2\sigma^2}\right) \quad (\text{A11})$$

is the Gaussian centered at μ . We use the same parameter values as those in Sec. II.

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