

Electrically Tunable Orbital Coupling and Quantum Light Emission from O-Band Quantum Dot Molecules

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We present the observation of electrically tunable quantum coupling of orbital states in individual InAs/InGaAs quantum dot molecules emitting in the telecom O-band ($\sim 1.3 \mu\text{m}$). By tuning the static electric field along the growth axis of the quantum dot (QD)-molecule, we observe pronounced anticrossings between excitonic transitions and determine the dependence of the interdot electron tunnel coupling on the interdot separation. As the electric field applied along the growth axis of the QD-molecules increases, positively charged exciton complexes sequentially emerge in the time-integrated emission spectra due to electrons escaping from the system while holes remain trapped. Moreover, for strong pumping, biexciton emission from the O-band molecules is identified. We demonstrate single-photon emission from the InAs/InGaAs QD-molecule emitting around $1.3 \mu\text{m}$ with a $g^{(2)}(0) = 0.017 \pm 0.002$ and explore the impact of tuning orbital coupling on the second-order correlation function.

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I. INTRODUCTION

The development of single-photon sources (SPSs) based on semiconductor quantum dot (QD) and QD-molecule (QDM) nanostructures emitting in the telecommunications bands is essential for integration with fiber optics and scalable silicon nanophotonic platforms [1–15]. Entangled multi-photon states are a key resource for measurement-based photonic quantum communication and computation [16–19]. In contrast to single QDs, it has been theoretically shown that vertically stacked QDMs hosting two optically addressable spins can deterministically generate

multi-photon states with the required two-dimensional entanglement structures [18].

Most state-of-the-art semiconductor-based SPSs are based on near-infrared (NIR) InAs QDs and QDMs grown on GaAs substrates by molecular beam epitaxy (MBE) or metal-organic vapor-phase epitaxy [20–30]. The vertical stacking of optically active NIR InAs/GaAs QDMs emitting at wavelengths $< 980 \text{ nm}$ (1.265 eV) has been carefully studied by many groups [31–33]. In this self-assembled QDM system, the lower QD acts as a stressor that defines preferential nucleation sites for the upper dot [31]. For conventional InAs/GaAs QDMs, the two QD layers are typically separated by a GaAs or AlGaAs barrier up to 13 nm -thick [23–29]. The first experimental demonstration of electrically tunable orbital coupling in the InAs/GaAs QDMs was demonstrated by Krenner *et al.* [34,35]. Vertical alignment of the two QDs and the strength of the tunnel coupling are governed by the thickness and Al-content of the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ layer separating them [23,24,27,34].

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The low-temperature (LT) emission wavelength of SPSs based on InAs/GaAs QDs and QDMs typically does not exceed $1.2\ \mu\text{m}$ ($1.033\ \text{eV}$) [36], limiting the compatibility of this quantum-emitter platform with standard silicon-compatible integrated photonic circuits and long-distance fiber-optic infrastructure. Therefore, further progress toward realizing scalable quantum technologies requires SPS operation in the low-loss telecom O- and C-bands, centered around 1.3 and $1.55\ \mu\text{m}$, respectively [1,2,8,19]. Promising platforms for telecom quantum light emission include InAs QD and QDM heterostructures grown on either GaAs or InP substrates [5,6,8–15] to enable: photonic-based prototyping of large-scale quantum key distribution [10], long-distance quantum transduction [27], quantum teleportation [9], and quantum computation protocols [10]. Both platforms are sufficiently mature to provide broad opportunities for enhancing photon extraction efficiency and boosting entanglement fidelities through the integration of InAs QDs and QDMs into photonic resonators [1,8,19]. Although deterministic and highly indistinguishable C-band SPSs based on InAs/InGaAlAs QDs grown on InP substrates have been demonstrated by Hauser *et al.* [19], reaching a raw two-photon interference visibility of up to $0.917(2)$, further progress toward electrically controlled QD-based SPSs grown on InP remains challenging [4,11,37,38]. In contrast to InAs QDs grown on InP substrates, which intrinsically emit in the O- and C-bands, redshifting the fundamental excitonic transition of InAs QDs grown on GaAs substrates into the O-band from ~ 1.2 to $\sim 1.3\ \mu\text{m}$ can be achieved by applying a strain-reducing layer (SRL) approach developed by Ustinov *et al.* [39]. The standard SRL approach, widely used for O-band data-transmission devices such as vertical-cavity surface-emitting lasers [36] and mode-locked comb lasers [40] operating at room temperature, consists of overgrowing self-assembled InAs QDs with a thin ($< 8\ \text{nm}$) In(Ga,Al)As(N) [41–43] alloy before completing the GaAs cap. The introduced SRL layer partially relaxes the strain field around the InAs QDs, named further InAs/InGaAs QDs, and modifies the confinement potential. This leads to a redshift of the excitonic transition [44] by $80\text{--}120\ \text{meV}$ while preserving compatibility with site-selective fabrication of SPSs [38,42,45] and integration with GaAs-based topological and conventional nanophotonic antennas or waveguides, including electrical control for charge-noise reduction [8,20–30,46]. Achieving not only waveguide integration but also the first electrically controlled, bright O-band SPS operating close to the fundamental coherence limit of InAs/InGaAs QDs has been recently demonstrated by Albrechtsen *et al.* [8]. These results highlight the potential of the O-band InAs/InGaAs QDs for scalable generation of cluster states at silicon-compatible wavelengths.

Here, we report the first synthesis and direct observation of electrically tunable orbital quantum coupling in O-band

QDMs formed by vertically stacking pairs of InAs/InGaAs QDs separated by a thin GaAs tunnel barrier. We demonstrate a high-quality MBE growth strategy for optically active O-band QDMs with a surface density $\leq 1\ \text{QDM}/\mu\text{m}^2$ and systematically correlate their structural and optical properties, thereby mapping the parameter space relevant for deterministic charging [25,29] required for the realization of a singlet-triplet ($S - T_0$) qubit [18] in telecom O-band. Consequently, we optimize the SRL thickness to ensure vertical correlation of the QD layers and investigate the range over which the tunnel-coupling strength can be tuned. To facilitate wide tunability of the exciton energy and orbital coupling via the dc Stark effect [47], we surround the O-band QDMs by AlGaAs barriers (see Fig. 1) and embed them into the intrinsic region of a vertical p-i-n diode. Coupling of orbital states within the QDM is evidenced by pronounced anticrossings in the optical spectra, arising from electron tunneling between the QDs. A progressive transfer of spectral weight toward increasingly *positively* charged excitonic complexes is observed with increasing electric field, indicating hole accumulation in the upper QD of the molecule. These signatures are reproducibly observed across multiple QDMs and for samples with different interdot separations. Our results establish clear structure-property relationships for O-band InAs QDMs incorporating an InGaAs SRL. Finally, we demonstrate quantum light emission around $1.3\ \mu\text{m}$ originating from coupled excitonic states of the QDM and single-photon emission with a $g^{(2)}(0) = 0.017 \pm 0.002$ under continuous-wave (cw) excitation.

II. EXPERIMENT

We investigated four samples grown by MBE: (i) a reference sample that contains only a single layer of InAs/InGaAs O-band QDs, and samples (ii), (iii), and (iv) containing two layers of QDs with interdot separations of 3 , 5 , and $10\ \text{nm}$ GaAs, respectively (see Fig. 1).

All samples were grown under identical conditions by MBE on undoped 2-inch GaAs(001) substrates using a Veeco Gen II MBE system. The growth rates were $2\ \text{\AA}/\text{s}$ for GaAs, $1\ \text{\AA}/\text{s}$ for AlAs, and $\sim 0.15\ \text{\AA}/\text{s}$ for InAs. The indium cell temperature was held constant during the growth of both the InAs QD layer and the $\text{In}_{0.25}\text{Ga}_{0.75}\text{As}$ SRL. A gradient deposition of 1.7 monolayer (ML) (nominal coverage at the wafer center) of InAs was performed in the sub-ML regime using a sequence of InAs growth and As exposure cycles with a growth time of $t_g = 3\ \text{s}$ and an interruption time of $t_b = 3\ \text{s}$ at $T_S = 520^\circ\text{C}$. The substrate was continuously rotated at $12\ \text{RPM}$ after the initial four sub-ML deposition cycles to establish a controlled InAs thickness gradient across the wafer. For all samples, the InAs QDs were grown on the nominally flat GaAs surface at

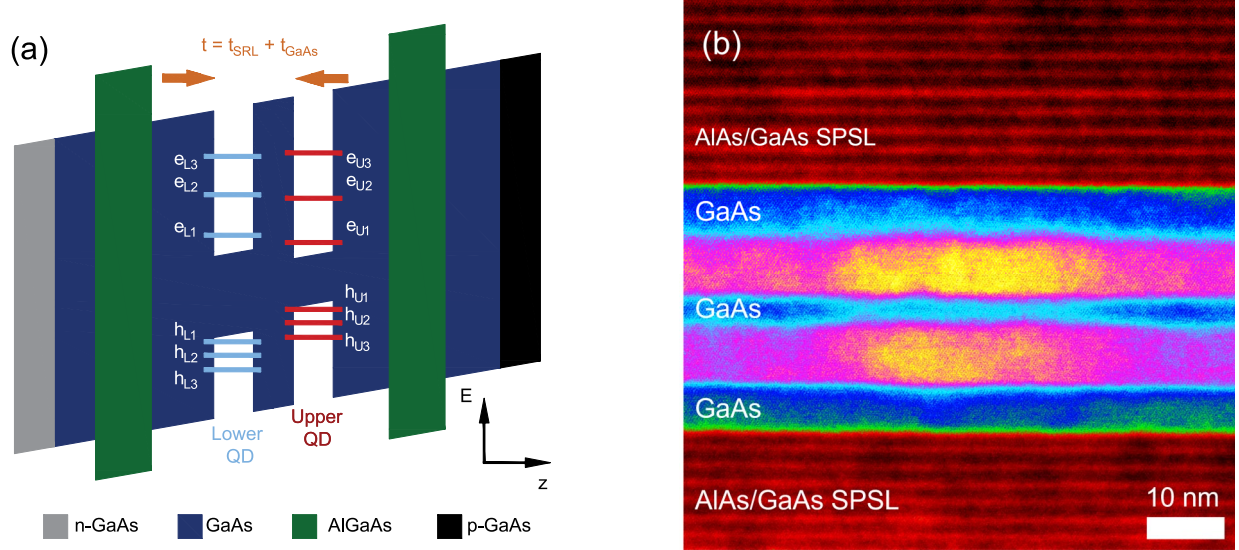


FIG. 1. (a) Schematic of p-i-n diode band diagram with InAs/InGaAs QDM including doped layers and AlGaAs barrier. (b) A false-color cross-sectional high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) image of a single InAs/InGaAs QDM with a 3 nm GaAs barrier.

$P_{\text{As}}(\text{BEP}) = 7 \times 10^{-6}$ Torr without a pattern-defining layer [48]. Following the growth of the first QD layer, T_s was reduced to 490 °C during a 90 s interruption under As flux before overgrowth with a nominally 7.5-nm-thick $\text{In}_{0.25}\text{Ga}_{0.75}\text{As}$ SRL where $P_{\text{As}}(\text{BEP})$ was increased to 1×10^{-5} Torr. After capping the first InAs/InGaAs QD layer with 2 nm of GaAs, T_s was linearly ramped back to 580 °C to deposit an additional 1–8 nm GaAs barrier layer, ensuring a smooth surface for subsequent growth of the second InAs/InGaAs QD layer.

As depicted schematically in Fig. 1(a), the InAs/InGaAs QDMs were embedded near the center of the p-i-n diode and surrounded by AlGaAs barriers, enabling electric-field tuning over the range 0–170 kV/cm while suppressing carrier tunneling escape from the QD(M) region [47]. The first InAs/InGaAs QD layer (lower) was deposited 5 nm above the 68-nm-thick barrier consisting of a short-period superlattice (SPSL) equivalent to $\text{Al}_{0.75}\text{Ga}_{0.25}\text{As}$ [42,47], while the second QD layer (upper) was capped with a 5 nm GaAs spacer followed by an identical SPSL barrier. The thickness of the undoped GaAs layers located between the p- and n-type GaAs contact layers and the SPSL barriers was varied to maintain the same total diode lengths. The undoped GaAs layer thickness of sample (i) is 85 nm. Figure 1(b) shows a representative high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) image of the QDM sample (ii), presented in a false-color filter to enhance contrast, revealing the SPSL barriers and the vertically stacked QDM.

Diode mesas with an area of $400 \times 400 \mu\text{m}^2$ were defined using maskless lithography and wet-etched down to the n-doped GaAs layer. Standard electrical contacts were fabricated by depositing a Ge/Au/Ni/Au metal

stack onto the n-GaAs, followed by annealing at 420 °C, and a Ti/Pt/Au metal stack onto the p-GaAs without an annealing step.

Micro-photoluminescence (μPL) measurements at LT were performed on all samples as a function of applied voltage (PLV) subject to non-resonant cw excitation at 895 nm. The emission from the InGaAs SRL was observed around 1000 nm at 4 K. For the statistical analysis of tunnel coupling energy (Δ_{tc}), we extended a scanning hyperspectral imaging technique (HSI) [3,49] also to enable voltage-dependent HSI imaging. Each scan yields a four-dimensional dataset comprising PLV spectra at each spatial position (x, y), providing statistics across multiple individual QDMs.

III. RESULTS AND DISCUSSION

Figure 2(a) shows typical PLV spectra recorded from the reference sample (i) with a single QD layer. These data were acquired from a region of the substrate having a density of $> 1 \text{ QD}/\mu\text{m}^2$. As expected, all emission lines 1–4 exhibit a pronounced quantum-confined Stark effect (QCSE) as the electric field is increased, characterized by

$$\Delta E_{\text{Stark}}^{\text{dir}} = E^{\text{dir}}(F) - E^{\text{dir}}(F = 0) = -p \cdot F - \beta \cdot F^2,$$

where F is the static electric field strength, p is the permanent exciton dipole moment, and β is the exciton polarizability [47,50]. The static electric field is $F = (V_{\text{bi}} - V)/d$, where $V_{\text{bi}} \approx 1.5 \text{ V}$ is the built-in voltage of the diode and $d = 334 \text{ nm}$ is the total thickness of the intrinsic region. The PLV data presented in Fig. 2(a) show a $> 10 \text{ meV}$ QCSE of two different InAs/InGaAs QDs with little quenching of the emission intensity due to the

AlGaAs barriers. Parabolic fits to the different transitions yield a permanent excitonic dipole moments of $p/e = 0.52$ nm and polarizabilities of $\beta = 0.27 \mu\text{eV kV}^{-2} \text{cm}^2$ for lines 3 and 4, and $p/e = 0.35$ nm and $\beta = 0.16 \mu\text{eV kV}^{-2} \text{cm}^2$ for the most intense lines 1 and 2, respectively. The exciton dipole moment p reflects the spatial separation between the centers of the electron and hole envelope wave functions in the absence of an external electric field. All four fitted bright emission lines, labeled 1–4, exhibit $p/e > 0$, indicating that the electron wave function is located below the hole wave function at $F = 0$ kV/cm, consistent with previous findings [51]. The polarizability is expected to be strongly dependent on the QD height [50] with $\beta \propto h_z^4$, giving rise to a substantially stronger parabolic Stark shift, by a factor up to 7, for InAs/InGaAs QDs compared to In(Ga)As/GaAs QDs.

Figures 2(b) and 2(d) present two representative anticrossings (ACs) observed in the weak excitation regime for the InAs/InGaAs QDM samples (ii) and (iii). As the applied voltage is tuned, the ACs emerge due to coupling

between spatially direct excitons (X_{dir}), where the electron-hole pair is confined within the same QD, and indirect excitons (X_{ind}), where the electron and hole reside in different QDs of the QDMs. X_{dir} follows the conventional QCSE, as described for sample (i), while the Stark shift of X_{ind} is given by $\Delta E_{\text{Stark}}^{\text{ind}} = e \cdot s_{\text{ind}} \cdot F$, where s_{ind} is the center-to-center interdot separation. This strong linear Stark shift enables tuning of X_{ind} into resonance with X_{dir} , resulting in an AC at the critical electric field F_{AC} due to electron tunnel coupling [34,52]. At the resonance, the electron (e) or hole (h) components of the excitonic wave function hybridize to form molecular-like states [34,52]:

$$E_{\pm} = E_1^0 + \frac{1}{2}\Delta \pm \frac{1}{2}\sqrt{\Delta^2 + (2t)^2}$$

with $\Delta = E_2^0 - E_1^0$ being the energy difference between the uncoupled states. Considering identical polarizabilities and intrinsic dipole moments, $\Delta = e \cdot p_{\text{ind}} \cdot (F - F_{\text{AC}})$, where $e \cdot p_{\text{ind}}$ represents the equivalent dipole moment of the X_{ind} . The dashed lines on Figs. 2(b) and 2(d) show fits

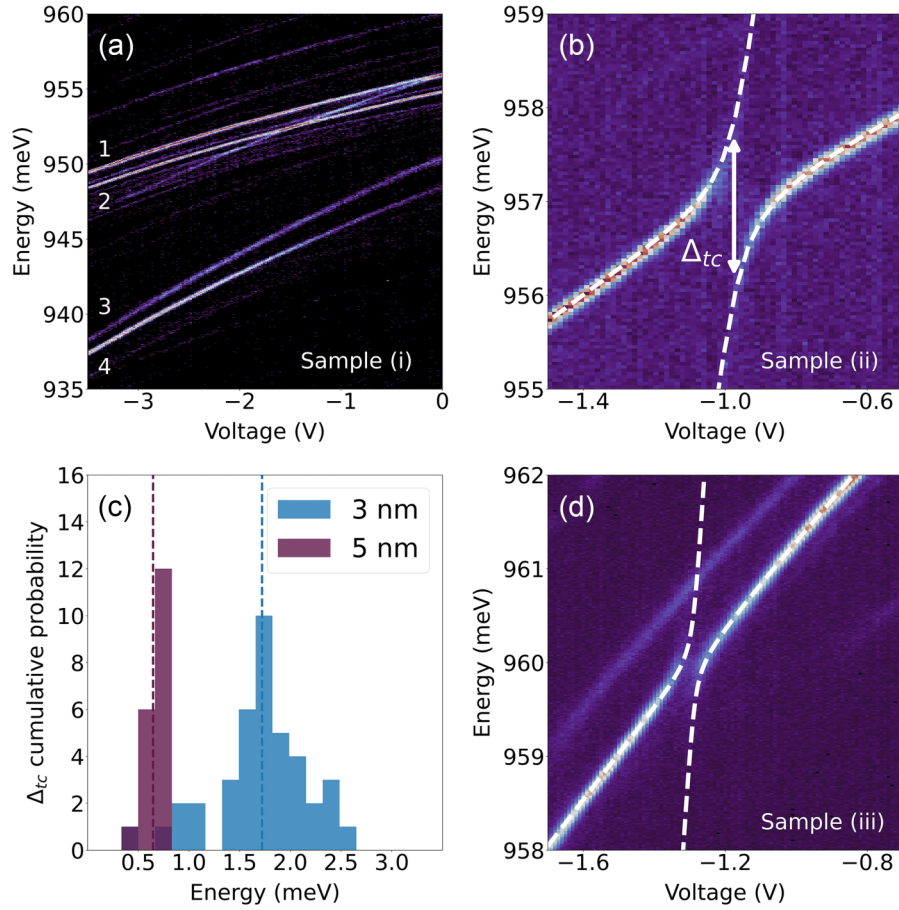


FIG. 2. (a), (b), and (d) PLV data recorder from a reference InAs/InGaAs QD sample (i) and InAs/InGaAs QDM samples (ii) and (iii) at $T = 4$ K under the cw 895 nm excitation with $P_{\text{exc}} = 1 \mu\text{W}$. (b), (d) The evolution of a representative anticrossing for GaAs barrier thickness t_{GaAs} of 3 and 5 nm, respectively. The anticrossing fit is shown as dashed lines. (c) Cumulative distributions of the tunnel-coupling energies Δ_{tc} for $t_{\text{GaAs}} = 3$ and 5 nm. The mean values of Δ_{tc} , 1.72 and 0.65 meV, are indicated by dashed lines.

to the data using the energy splitting of the anticrossing, which is then

$$\Delta_{tc} = \sqrt{[e \cdot p_{ind} \cdot (F - F_{AC})]^2 + (2t)^2}.$$

At resonance, the two hybridized excitons are split by a tunnel-coupling energy $\Delta_{tc} = 2t$, giving rise to the experimentally observed AC. In this regime, the direct and indirect excitons have identical oscillator strengths. We have determined Δ_{tc} by fitting both branches, subtracting the lower branch energy from the upper branch, and identifying the minimum splitting value.

Spin-spin exchange interactions in a charged QDM are primarily determined by Δ_{tc} [25,53], which can be tuned by varying the tunnel barrier thickness t_{GaAs} between the QD layers. To statistically investigate the dependence of Δ_{tc} on the interdot barrier thickness t_{GaAs} in our QDMs, we performed μ PL measurements on InAs/InGaAs QDM samples (ii), (iii), and (iv) with different $t_{GaAs} = 3, 5$, and 10 nm, respectively. For this, we used the state-of-the-art HSI technique [3] extended with a bias-voltage sweep at each scan step. The primary criterion for identifying an emitter as a QDM was the observation of the AC in the bias-dependent μ PL spectrum acquired at excitation power (P_{exc}) lower than 1 μ W to identify only AC related to X , which provides direct statistics of Δ_{tc} of orbital tunnel coupling as a function of t_{GaAs} . We further note that InAs coverage of the lower and upper QD layers is a critical parameter for the vertical alignment of QDMs grown in the gradient regime [44,48]. For QDM surface densities exceeding 1 emitter/ μm^2 , the stacking probability determined by STEM was found to approach 100% for samples (ii) and (iii). STEM measurements of sample (iv) were not carried out in this study. However, in regions with densities below 1 emitter/ μm^2 , the stacking probability can be lower than unity either due to differences in the nucleation dynamics of the lower and upper QD layers as a function of the local InAs coverage [44] or due to the increase of the interdot separation [31,54], that has been confirmed in our HSI measurements acquired in $20 \times 20 \mu m^2$ areas with the optical surface density lower than 0.2 emitters/ μm^2 . The full dataset contained around 60 quantum emitters for each sample (ii)–(iv). Based on these data, the probability of observing anticrossings was estimated to be 74% and 32% for the QDM samples with $t_{GaAs} = 3$ and 5 nm, respectively. No anticrossings could be observed for sample (iv) with $t_{GaAs} = 10$ nm, indicating that the vertical correlation between the pairs of QDs forming the molecules is lost. However, not every emitter showed emission within the telecom O-band, similar to the results of HSI for the O-band single QD layer sample reported in Ref. [44]. For our statistical analysis, only anticrossings observed within the O-band wavelength range of 1290–1310 nm were included in the statistics presented in Fig. 2(c).

The excluded emitters did not provide a statistically significant dataset for establishing correlations between Δ_{tc} and the emission wavelength. Therefore, 37 and 16 exciton anticrossings for samples (ii) and (iii), respectively, were included in the present statistical analysis. Anticrossings associated with other excitonic complexes were also excluded from the statistical analysis due to a more complex relation between Δ_{tc} and t than for X , as reported in Refs. [25,29,55,56]. As t_{GaAs} increases from 3 to 5 nm, the mean value of Δ_{tc} decreases from 1.72 ± 0.48 to 0.65 ± 0.1 meV, indicated by dashed lines in Fig. 2(c). The behavior of an excitonic two-particle wave function in a strained, symmetry-broken QDM system is intrinsically non-trivial [57]. As expected, the final value of Δ_{tc} in asymmetric QD pairs depends sensitively on the microscopic structure, including the relative QD size, In-composition profile, and strain distribution. The latter is strongly affected by the interdot separation and contributes to carrier confinement and, consequently, to Δ_{tc} in the QDM. We therefore attribute the larger standard deviation of Δ_{tc} observed for sample (ii) to the stronger influence of strain-related effects at smaller interdot separations. Nevertheless, within single- and many-particle descriptions of tunnel-coupled systems [57,58], the electron tunneling rate $T \propto \Delta_{tc}$ at the given applied electric field can still be qualitatively described as being mostly dependent on the height and width of the potential barrier separating the QDs. Consequently, an increase in either the interdot barrier height (e.g., via a higher Al content in the barrier separating the QDs) or the barrier thickness, as well as deeper localization, leads to a suppression of tunneling and, consequently, Δ_{tc} . The absence of ACs for a total lower wetting layer (WL) to upper WL separation of $t_{GaAs} + t_{SRL} = 17$ nm [see Fig. 1(a)] confirms that the tunnel coupling decreases sharply with increasing barrier thickness, occurring more rapidly in QDM systems with enhanced carrier confinement compared to the conventional NIR In(Ga)As/GaAs [27].

The panels (a) and (b) of Fig. 3 present PLV data recorded from sample (ii) in a broad range of applied bias when excitation is weak (300 nW) and strong (5 μ W), respectively. For weak excitation, the formation of multi-exciton species is suppressed, and the majority of the emission lines stem from single exciton species populating the QD-molecule. At low electric fields (< 70 kV/cm), a dominant spectral line, marked X , exhibits a QCSE similar to that of a single QD. In contrast to the single QD, as the electric field is tuned into the range 70 to 85 kV/cm, we observe AC, labeled AC1, in the spectrum. Fitting yields $\Delta E_{dir/ind}(F=0) = |E_0^{X_{ind}} - E_0^{X_{dir}}| \approx 74$ meV, along with $s_{ind} \approx 9.9$ nm, which is in agreement with the nominal WL-to-WL layer separation $t = t_{SRL} + t_{GaAs}$ of 10 nm. For electric fields $F > 85$ kV/cm, the electron-hole overlap for X_{ind} becomes negligible, and only the X_{dir} remains optically active in luminescence [see Fig. 3(a)].

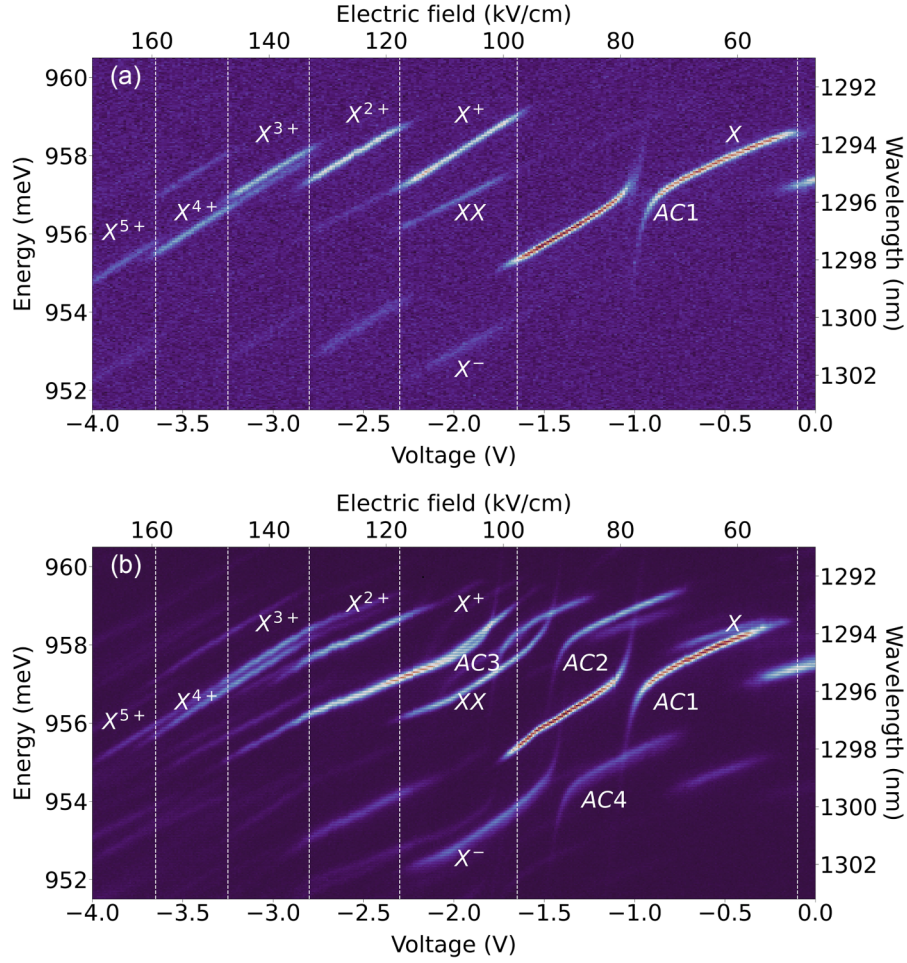


FIG. 3. PLV spectra of InAs/InGaAs QDM sample (ii) with $t_{\text{GaAs}} = 3$ nm acquired at $T = 4$ K under cw 895 nm excitation with $P_{\text{exc}} = 300$ nW (a) and 5 μ W (b), respectively.

As the electric field is further increased ≥ 98 kV/cm, a series of abrupt steps are observed in the emission energy at specific voltages, indicating that two layers of QDs become tunnel-coupled, and charge transfer occurs over time scales faster than the radiative lifetime. They are denoted in Fig. 3(a) by the vertical dashed lines on the figure. At these steps, the dominant QDM emission lines quench and are sequentially replaced by other lines primarily shifted to higher energy. This characteristic behavior is indicative of sequential charging of the QDM and the dominant emission shifting to different charge states of the system. We note that optical excitation always generates equal numbers of photoexcited electrons and holes. As such, emission from an effectively positively charged complex in the molecule requires that electrons transfer out of the QD, being trapped against the lower AlGaAs barrier. We label the emission in these regions X^{n+} , where $n = 1 \rightarrow 5$ and tentatively identify the different features as arising from effectively *positively* charged excitons in the upper QD of the molecule. Hereby, the axial electric field results in preferential hole transfer into the

upper QD and the faster comparative tunneling escape results in hole accumulation.

We now support our identification of these features. As shown by Regelman *et al.* [59], the relative energy of charged and neutral exciton complexes reflects the interplay between: (1) the total exciton energy expressed as $\Delta E_X = E_{e1} - E_{h1} - J_{e1h1}$, where E_{e1} (E_{h1}) are the lowest (highest) orbital-state energies of the electron (hole), and J_{e1h1} is the electron-hole pair binding energy; (2) The *Coulomb shift* (V_{ij} , with $i, j = e, h$) accounting for the difference between Coulomb repulsion and attraction terms between the recombining exciton and the spectator carriers; (3) The always negative *exchange shift* (ΔE_{exch}) responsible for the appearance of multiple spectral lines within the same voltage range. Open shells in the final state give rise to spin multiplets whose energies depend on the relative spin orientation of the confined carriers (e.g., the range less than -2.3 V in Fig. 3), where the doublet structure X^{2+} results from different triplet or singlet final states [60] and (4) The *correlation shift* (ΔE_{corr}) arising from variations in carrier-carrier correlations within the

interacting multicarrier system confined in the QD. According to Coulomb interaction considerations, the electron wave function is more weakly localized than that of the hole due to its smaller effective mass. Therefore, the relative strength of Coulomb interactions scales as $|V_{ee}| < |V_{eh}| < |V_{hh}|$. Assuming a harmonic confinement potential and accounting for the effective masses, $|V_{hh}| + |V_{ee}| > 2|V_{eh}|$ [50,61]. Based on these considerations, the negatively charged exciton (X^-) is expected to exhibit a redshift, whereas the positively charged exciton (X^+) shows a blueshift relative to the neutral exciton (X). Furthermore, the magnitude of the X^- shift is predicted to be smaller than that of X^+ in InAs/GaAs QDs [61].

Taking the brightest line in each bias window, the experimentally observed energy shifts are:

$$\begin{aligned} X^- - X &\approx +3.65 \text{ meV}, \\ X^{2+} - X^+ &\approx +1.5 \text{ meV}, \\ X^{3+} - X^{2+} &\approx +0.85 \text{ meV}, \\ X^{4+} - X^{3+} &\approx -0.2 \text{ meV}, \\ X^{5+} - X^{4+} &\approx +0.2 \text{ meV}. \end{aligned}$$

These values are qualitatively in agreement with the typical values reported for In(Ga)As/GaAs QDs [59,60]. All our samples studied here were grown under identical growth conditions for the upper and lower QD layers, which almost unavoidably results in slightly larger upper QD with higher In content compared to the lower QD, as confirmed by STEM [see Fig. 1(b)]. Consequently, the upper QD has stronger carrier confinement and polarizability in the coupled asymmetric QDM. Thus, we expect that at 0 V bias the electron/hole energy states satisfy $E_{e1}^{\text{lower}} > E_{e1}^{\text{upper}}$, while $E_{h1}^{\text{lower}} < E_{h1}^{\text{upper}}$ at $F < F_{AC1}^X$, as well as $\beta_{QD}^{\text{upper}} > \beta_{QD}^{\text{lower}}$ [see Fig. 1(a)]. The observed charging behavior in Fig. 3(a) for the InAs/InGaAs QDM is in good agreement with the charging diagram of large, In-rich InAs/GaAs QDs emitting around 1.2 μm and embedded in an n - i -Schottky diode [61]. This correspondence further supports the picture that the lower InAs/InGaAs QD acts as a carrier reservoir for the upper QD. Moreover, the SRL overgrowth induces a redshift of the electron and hole states of InAs QD emission without significantly altering the hole-level splitting. At higher reverse bias, after reaching the neutral exciton anticrossing $F > F_{AC1}^X$ and $E_{e1}^{\text{lower}} < E_{e1}^{\text{upper}}$, while still $E_{h1}^{\text{lower}} < E_{h1}^{\text{upper}}$. Therefore, the positively charged complexes X^{n+} blueshifted states are likely formed optically through the generation of an initial X or biexciton (XX) in the upper QD, followed by electron (hole) tunneling to the lower (upper) QD.

Figure 3(b) shows high-power PLV data. Increasing the excitation level leads to a pronounced modification of the PLV spectrum of the InAs/InGaAs QDM for electric fields $> 80 \text{ kV/cm}$. Most strikingly, the emission lines exhibit

abrupt discrete steps in the X^+ -marked PLV region shown in Fig. 3(a) and evolve into pronounced features displaying additional anticrossings, labelled AC2-4 on the figure. Small deviations from the parabolic Stark-shift behavior of the direct excitonic transitions become visible in this regime in Fig. 3(b), for example, for the X emission line at voltages below -1.5 V , near the switching regions marked by dashed lines. Following the analysis reported by Dalgarno *et al.* [62] carried out for the many-body exciton states in QD coupled to a Fermi sea, we attribute these signatures to optically induced weak hybridization between populated orbital states of the two QDs. We note that such optical signatures were not observed in the μPL measurements of O-band QDMs with a 5-nm-thick interdot GaAs, consistent with weaker Coulomb interactions at increased interdot separation. Different charge states exhibit transitions with anticrossings that occur at different electric fields [55]. Anticrossings are not only limited to resonances between the lowest energy orbital states, but they can be used to optically probe the orbital states in each QD forming QDM. However, we note that the specific PLV anticrossing pattern of an excitonic state depends on couplings that occur in both initial and final states [55]. For example, the characteristic “X-shaped” anticrossing pattern that typically occurs for a trion ($X^\pm$) in a QDM [35,52,55,56] is not observed if the initial and final states of the optical transition do not undergo resonance at similar electric fields. When the upper and lower QDs have significantly different confinement energies, characteristic X-patterns are not observed for X^n states, depending on the preferential pathways of carrier relaxation and tunneling escape rates [56]. We note that the slope $p_{\text{ind}} = \Delta E / \Delta F$ has the same sign for AC1–AC4, indicating that the delocalization of the same component of the exciton wave function is responsible for all tunnel couplings.

We mapped the specific electric fields at which different ACs occur to the change of a few particle Coulomb interactions. Hereby, we assume that the differences between the resonance fields F_{AC_i} (with $i = 1\text{--}4$) arise from Coulomb interactions between various orbital states, and express the shift of F_{AC_i} relative to F_{AC_1} to an energy offset using

$$\phi_{1i} = p_{\text{ind}}(F_{AC_i} - F_{AC_1}),$$

where $p_{\text{ind}} = e s_{\text{ind}} \approx e \cdot 9.9 \text{ nm}$.

The results of this analysis are summarized in Table I. We find that the tunnel coupling energies are similar for all anticrossings. As shown in Ref. [63], the $s - p$ orbital splitting for electrons is expected to be considerably larger than for holes. The electron $s - p$ splitting is typically several tens of meV, whereas for holes it is expected to be $\leq 10 \text{ meV}$. The $p - d$ splitting exhibits the opposite behavior: For holes, the magnitude of the splitting remains

TABLE I. Summary of tunnel-coupling parameters.

AC #	Δ_{tc} (meV)	F_{AC} (kV/cm)	ϕ_{li} (meV)
AC1	1.78	76.1	0
AC2	2.1	91.6	15.3
AC3	1.74	101.9	25.5
AC4	1.8	90.4	14.0

nearly constant at approximately 10 meV, while for electrons it changes nonmonotonically with height. The wave functions of the electron states are similar in shape to the conventional s -, p -, and d -like orbitals, whereas the hole states cannot be classified in this manner due to strong interband mixing [63]. Consequently, coupling between two electron states with different orbital characters, such as $s - p$ or $s - d$, can occur through partial overlap of their in-plane wave functions belonging to different orbitally excited states [64]. However, such couplings are expected to exhibit reduced Δ_{tc} compared to, e.g., $s - s$ or $p - p$ coupling.

The power dependence of the integrated μ PL intensity, $I \propto P_{exc}^\alpha$, measured in the range $-1.2 \text{ V} \rightarrow -1.3 \text{ V}$ reveals that the exponent α for AC1 is 0.95 ± 0.01 , supporting our attribution of it as arising from a neutral exciton. In contrast, the exponents associated with AC2, AC3, and AC4 are 2.19 ± 0.04 , 1.55 ± 0.09 , and 1.59 ± 0.08 , respectively. These values are indicative that the associated transitions are biexciton XX and trion ($X^\pm$) states, respectively. A definitive interpretation of the nature of the multi-particle anticrossings cannot be made since shell filling effects become important as the excitation density increases toward the exciton saturation level P_{sat}^X , which occurs at a power 16.7 times higher than that in Fig. 3(a). Such shell filling effects are further enhanced by the presence of AlGaAs barriers [see Fig. 1(a)], which suppress carrier tunneling out of the QDM. Moreover, when

the lowest energy orbital states energies become resonant at $E_{e1}^{lower} < E_{e1}^{upper}$ but $E_{e2}^{lower} > E_{e2}^{upper}$, the optical formation of X^{n+} states from X or XX in the upper QD, followed by electron tunneling to the lower QD, becomes less straightforward, while hole transfer into the upper QD becomes more likely for $F > F_{AC1}$. Therefore, AC2–AC4 likely originate from a different coupling mechanism within the QDM, namely the Coulomb-mediated hybridization of excitonic states, described by Ardelt *et al.* [65]. At these resonances, both the electron and hole components of the excitonic complexes are hybridized across two single-particle orbitals. Such ACs have been shown to involve different excitonic states $|\psi_i\rangle$ having a unique configuration of single particle states and occur whenever a direct excitonic complex is tuned into resonance with an indirect one with $\Delta_{tc} \propto 2 \cdot t \cdot |\langle \psi_{dir} | \psi_{ind} \rangle|^2$, even in the absence of resonant single particle tunnelling. In the present case, after charge transfer between the two QDs, the stronger Coulomb binding energy of the resulting direct exciton state requires an additional electric field to restore resonance between the direct and indirect states, thereby forming the AC.

Based on the power dependence of the integrated μ PL intensity, we tentatively identify AC2 as being a biexciton XX transition in the upper QD. The origin of AC4 at $F > F_{AC1}^X$ and $E_{e1}^{lower} < E_{e1}^{upper}$ could be partially explained by the involvement of the hot negatively charged trion (X^-) in the upper QD [66], with configuration $1e_s 1e_p 1h_s \rightarrow 1e_p$, where e/h denote electron/hole states in the s - or p -levels, respectively. The redshifted AC4 in Fig. 3(b) exhibits features absent in AC1–AC3. We plot a representative μ PL spectrum of the InAs/InGaAs QDM sample (ii) in Fig. 4(a), acquired at an applied bias of -1.2 V under cw 895 nm excitation (above the SRLs) with $P_{exc} = 5 \mu\text{W}$. The metastable hot X^- involves spin-triplet and spin-singlet configurations [66–68], resulting in a characteristic emission-line doublet in the PLV spectra.

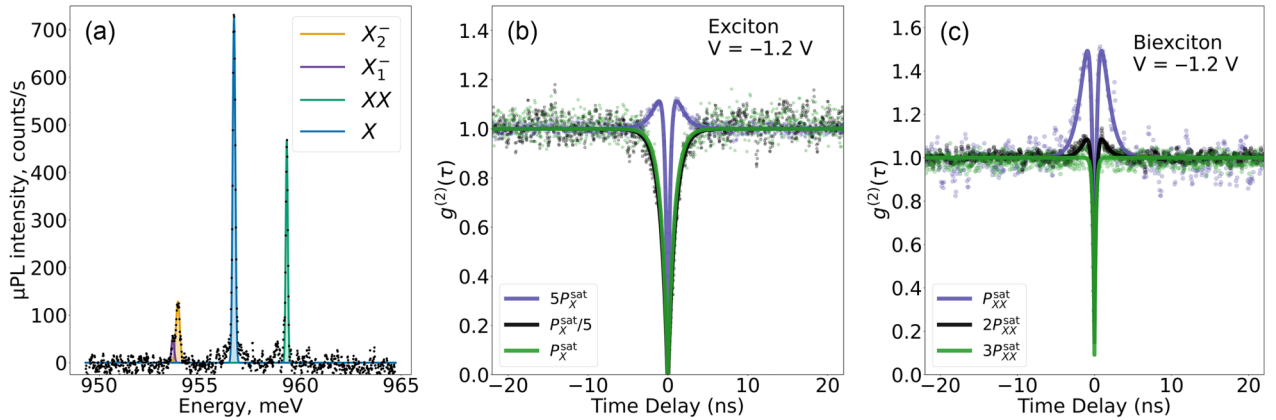


FIG. 4. (a) μ PL spectrum of the InAs/InGaAs QDM sample (ii) acquired at an applied bias of -1.2 V under cw 895 nm excitation (above the SRLs) with $P_{exc} = P_{sat}^X$. Power-dependent second-order autocorrelation measurements and corresponding fits for the exciton (b) and biexciton (c) lines of sample (ii), recorded under 895 nm cw excitation at a bias of -1.2 V .

When tunneling escape of the p -shell electron is suppressed, the hot-trion lifetime becomes sufficiently long for radiative recombination to occur, since relaxation from the electron spin-triplet configuration to the singlet ground-state trion is spin-forbidden and therefore proceeds via a comparatively slow spin-relaxation process [66]. We label the lower- and higher-energy lines as X_1^- and X_2^- , respectively. The lower-energy X_1^- transition, which exhibits predominantly direct character in the 70–85 and 95–120 kV/cm ranges, vanishes near $F_{AC4} = 90.4$ kV/cm [see Fig. 3(b)]. Simultaneously, the energy splitting between X_1^- and X_2^- decreases monotonically from 120(20) μ eV and vanishes as the two transitions approach AC4. In contrast, the positively charged trion X^+ , with the spin singlet configuration $2h_s, 1e_s \rightarrow 1h_s$, contributes to AC3 without exhibiting an analogous doublet emission structure. The observed differences $\phi_{12} - \phi_{14} \approx 1.3$ meV and $\phi_{12} - \phi_{13} \approx 10.2$ meV are consistent with the variations in relative Coulomb interaction energies among these excitonic configurations.

To confirm that AC2-4 are indeed related to the charged excitonic states, we studied the power dependence of the second-order $g^{(2)}(\tau)$ intensity autocorrelation of direct excitonic states for voltages around AC1 and AC2 under non-resonant cw excitation at 895 nm. For these measurements, the voltage was fixed at -1.2 V to avoid a change in tunneling rate. This regime corresponds to the $E_{e1}^{\text{lower}} < E_{e1}^{\text{upper}} < E_{e2}^{\text{lower}}$ and $E_{h1}^{\text{lower}} < E_{h1}^{\text{upper}}$ level alignment. Distinguishing between different excitonic charge states using autocorrelation measurements can be carried out based on the character of correlation spectra, which is unique to each complex [69]. Figure 4(b) exhibits the expected excitation rate-mediated single-photon behavior of a two-level system (InAs/InGaAs QDM) of the X state emitting at 956.7 meV, described, in the general case, by

$$g^{(2)}(t) = (1 - Ae^{-t/\tau_a}) \cdot (1 + Be^{-t/\tau_b}),$$

where B and τ_b are set to zero in the absence of bunching. The fitting parameters $g^{(2)}(0), \tau_a, \tau_b$ are summarized in Table II.

Table II shows that the extracted τ_a times for the X and XX transitions are 0.89(1) and 0.50(13) ns at the

TABLE II. $g^{(2)}(t)$ fitting parameters.

Power	$g^{(2)}(0)$	τ_a (ns)	τ_b (ns)
$P_X^{\text{sat}}/5$	0.04(4)	1.1(1)	0
P_X^{sat}	0.017(2)	0.89(1)	0
$5P_X^{\text{sat}}$	0.010(2)	0.28(1)	2.2(1)
P_{XX}^{sat}	0.18(8)	0.50(13)	1.23(17)
$2P_{XX}^{\text{sat}}$	0.13(2)	0.28(2)	1.09(14)
$3P_{XX}^{\text{sat}}$	0.09(2)	0.18(1)	0

corresponding P^{sat} , respectively. The value for X is in good agreement with non-cavity-enhanced telecom InAs/InGaAs QDs exciton lifetimes of $\tau_{\text{rad}}^X \approx 1$ ns reported previously [8,45]. The ratio $\tau_a^X(\text{sat})/\tau_a^{XX}(\text{sat}) \approx 1.8$ is close to the expected 2, reflecting the double probability of radiative recombination for two electron-hole pairs in XX [70] and is in good agreement with the α values determined from the power dependence of the integrated μ PL intensity in this study.

As long as the X state does not reach intensity saturation [green and black experimental data in Fig. 4(b)], the correlation spectra exhibit a pronounced antibunching dip down to $g^{(2)}(0) = 0.017(2)$, with a width determined by the exciton lifetime. With increasing excitation power density, additional bunching at short time delays appears (lavender data and fit curve), decaying toward the average coincidence level over a timescale longer than that expected from radiative lifetime τ_{rad}^X . Since the X transition τ_{rad}^X and the corresponding radiative recombination rate R_{rad}^X are not expected to depend on excitation power at a fixed dc bias, the decrease of the antibunching time constant τ_a^X with increasing P_{exc} indicates an increase of the effective pump rate R_{pump}^X . A pronounced antibunching dip is observed in both regimes: when $R_{\text{rad}}^X/R_{\text{pump}}^X > 1$, where τ_a^X is mainly limited by τ_{rad}^X , and when the average steady-state occupation probability of the upper QD is larger. Consequently, the probability of observing only a single electron-hole pair is reduced, and the X -line emission exhibits bunching caused by carrier shelving or Auger-type processes [69,71]. We attribute the magnitude of this bunching and the associated decay time τ_b to the X recovery time in the charge configuration of the QD or QDM. The excitation rate-mediated single-photon behavior of cw $g^{(2)}(\tau)$ presented in Fig. 4(b) supports the assignment of AC1 to the anticrossing of the neutral exciton X and the upper QD at -1.2 V bias is tuned into a regime similar to a single QD sample, i.e., the $E_{e1}^{\text{lower}} < E_{e1}^{\text{upper}} < E_{e2}^{\text{lower}}$ and $E_{h1}^{\text{lower}} < E_{h1}^{\text{upper}}$ level alignment. The extracted value of $\tau_b^X = 2.2(1)$ ns at the corresponding excitation power of $5P_X^{\text{sat}}$ may be attributed to the onset of a biexciton emission in this excitation regime with the doubled probability of radiative recombination.

The biexciton autocorrelation function of the line associated with the AC2 exhibits a markedly different behavior compared to the power dependence of the X state. In this case, a strong characteristic bunching is observed at short time delays under weak cw excitation for the line at 958.5 meV, as presented in Fig. 4(c). The width of the dip at P_{exc}^{XX} confirms the fast decay rate of XX . An analysis of the XX dynamics of the upper QD and the results of the cw $g_2(t)$ measurements at a bias of -1.2 V can be carried out using a standard three-level rate model. In such a regime, immediately after a cascade emission of a biexciton photon, the upper QD is left with one e - h pair, which

may either decay radiatively to the ground state or capture an additional carrier or an e - h pair to form $X^\pm$ or XX [69,72]. At relatively low P_{exc} , the cascade XX transition radiatively decays to the X state. If this decay is followed by XX reexcitation, the $\tau_b^{XX}(P_{XX}^{\text{sat}})$ bunching time is expected to occur on a timescale comparable to the $\tau_a^X(P_X^{\text{sat}})$ [72], which is in good agreement with the determined parameters in Table II. As P_{exc} increases from P_{XX}^{sat} to $3 P_{XX}^{\text{sat}}$ using 895 nm cw excitation, a gradual reduction of the bunching amplitude is observed within the measured time-delay window in Fig. 4(c). This behavior reflects the power-dependent pumping dynamics of the biexciton state. Simultaneously, the fitted antibunching time decreases from approximately 0.50(13) to 0.18(1) ns. We attribute this reduction in τ_a to an increasing contribution from free-carrier density around QDM, accelerating the XX pumping rate. Under stronger non-resonant cw excitation, we have observed the reduction of bunching decays, which effectively narrows the XX antibunching dip in the upper QD.

The achievement of electrically controlled orbital tunnel coupling in O-band QDMs offers wide opportunities not only for controlling single-photon emission demonstrated for near-infrared QDMs [26], but also for developing deterministic optical charging protocols in coupled QDMs within the telecom spectral range [18]. Considering recent progress in O-band SPSs operating close to the InAs/InGaAs QD fundamental coherence limit demonstrated by Albrechtsen *et al.* [8], we carry out a linewidth analysis of the developed O-band QDMs. The QDM emission shown in Fig. 4(a) was detected using a spectrometer with a focal length of 750 mm, equipped with a 1200 lines/mm grating and an InGaAs linear array detector. The spectral resolution of the μPL setup is ~ 22 – 25 μeV . Although the extracted τ_a of the upper-QD exciton measured at P_X^{sat} without cavity enhancement is close to 1 ns, the full width at half maximum of the excitonic emission lines $\Gamma^X(\mu\text{PL})$ observed in samples (ii)–(iv) was not found to be lower than 100 μeV . This value is approximately a factor of 5–6 larger than $\Gamma^X(\mu\text{PL})$ measured for non-electrically controlled samples containing a single InAs/InGaAs QD layer embedded in a GaAs matrix, as demonstrated in our previous study [44]. It is comparable to the linewidths of O-band InAs/InGaAs QDs reported by Olbrich *et al.* [71], but remains significantly broader than the near-Fourier-transform-limited linewidths reported for single QDs [8,73].

We attribute the broad $\Gamma^X(\mu\text{PL})$ to the significant contribution of charge noise in the environment of the presented O-band QDM samples. The QD emission lines are sensitive to the single-charge level fluctuation in the local environment [30,73]. The AlGaAs barriers implemented in our heterostructure design not only extend the electric-field tunability range, but can also enhance local charge fluctuations due to suppressed carrier tunneling.

Similar effects were previously identified in resonance fluorescence measurements of O-band InAs/InGaAs QDs by Al-Khuzheyri *et al.* [74]. To minimize charge noise in the environment of O-band QD(M)-based active-region heterostructures containing high AlGaAs barriers, two strategies can be considered: (1) modification of the voltage-sweep protocol during PLV measurements, following Heiss *et al.* [75], and (2) further optimization of the epitaxial growth procedure, as demonstrated by Albrechtsen *et al.* [8]. In the present work, a standard dc bias was applied to all studied diodes in order to demonstrate orbital tunnel coupling in O-band QDMs. Following the first strategy, future PLV measurements should incorporate a reset-voltage pulse into the sweeping procedure, similar to $e(h)$ -storage QD(M) heterostructure designs, in which an AlGaAs barrier is embedded only on one side of the QD(M). Such a reset pulse can suppress charge accumulation and, consequently, reduce charge noise, as demonstrated previously for NIR InAs/GaAs QDMs [25,29,66]. The second strategy, epitaxial optimization of O-band QDs and QDMs, requires addressing two potential sources of charge noise: (a) impurities in aluminum-containing layers embedded in the QD(M) active region, as discussed by Nguyen *et al.* [76], and (b) charge-carrier traps associated with the strain-reducing layers, as described by Olbrich *et al.* [71]. Recent progress on silicon-compatible O-band QD-based single-photon sources [8] confirms that InAs QDs overgrown with an InGaAs SRL can operate close to the fundamental coherence limit. This motivates the further development and integration of QDM-based SPSs hosting two optically addressable spins, with the long-term goal of realizing theoretically proposed two-dimensional photonic cluster states in the telecom O-band [18].

IV. CONCLUSION

In summary, we have demonstrated the electrically tunable quantum coupling of orbital states in individual InAs/InGaAs quantum dot molecules emitting around 1.3 μm . By tuning the applied electric field, we observed anticrossings of distinct excitonic transitions in a series of InAs/InGaAs QDM samples with $t_{\text{GaAs}} = 3$ and 5 nm, using both HSI and standard μPL techniques. The developed heterostructure design provides a platform for optical initialization, coherent manipulation, and readout of spin states in charged QDMs operating in the telecom O-band. Under strong excitation, biexciton emission from the O-band QDMs is identified. Finally, we have investigated the single-photon emission from InAs/InGaAs QDMs.

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The authors have no conflicts to disclose.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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