Supplementary Materials for

On-chip scalable highly pure and indistinguishable single-photon sources in ordered arrays: Path to quantum optical circuits

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References
1. SESRE: Spatially ordered and spectrally uniform MTSQDs

Of the competing on-chip solid state quantum emitters, the self-assembled quantum dots (SAQDs), being on-demand source, (and owing to the ease of synthesis) have been studied the most (8) but, given the underlying fundamental physics of lattice mismatch strain-driven spontaneous formation of the defect-free 3D islands during vapor phase deposition (64, 65) are random in location and nonuniform in size, shape, and composition that manifests in large inhomogeneity (~50nm) in spectral emission in the most commonly employed system of InGaAs on GaAs (001) substrate (65). This has severely limited all demonstrations of on-chip SAQD-cavity-waveguide integrated structures to a single SAQD (found through search over a large number) with a few exceptions that employ structures containing two SAQDs (50). The need for the number of physical qubits for practical systems for quantum information processing is estimated to range from a few hundred (for simplest quantum chemistry simulations) to millions (for computation) of single photon sources (SPSs) (66). To overcome this severe limitation, various approaches have been taken to induce site-selective formation of quantum dots (21-25, 67, 68) utilizing chemically and structurally pre-patterned substrates. Interested readers can find pertinent information in references (25), (67), and (68). While each of these approaches has its merits, none of them meet simultaneously all the strict requirements of (i) having one quantum dot per site in a configuration compatible with on-chip integration (i.e. planar / horizontal) architecture, (ii) the photons having sufficiently uniform spectral properties across a scalable array, and (iii) applicable to a wide variety of material combinations thereby covering a wide range of spectral emission from the ultraviolet to visible, and near-infrared to mid- and long wavelength infrared regimes. An exception has been the approach of substrate-encoded size-reducing epitaxy (SESRE) (16, 15, 26) briefly recalled here.

The MTSQDs reported in this manuscript are synthesized using the SESRE approach (16) that exploits growth on designed non-planar patterned substrates, i.e. patterned structurally such that the tailored surface curvature induces surface stress gradients (capillarity) that direct adatoms during deposition preferentially to mesa tops (16,15,26) for selective incorporation through control on the relative kinetics of adatom incorporation on the contiguous facets present in the designed curvature. For the (001) surface oriented substrates of the tetrahedrally-bonded semiconductors of groups IV, III-V, and II-VI, the <100> edge orientations of square mesas provide four-fold symmetry and thus potentially symmetric migration of adatoms from the sidewalls to the top (Fig. S1(a)). The preferential incorporation at the mesa-top leads to growth-controlled mesa size reduction (Fig. S1(b)), enabling in-situ preparation of contamination-free and defect-free nanomesa of the desired size utilizing homoepitaxy under controlled growth kinetics. A quantum dot (QD) can be formed on the size-reduced nanomesa with crystallographic controlled size-wall and controlled base length and thickness through heteroepitaxy. Thus, SESRE can be used to synthesize QD in spatially ordered arrays with growth controlled QD size and shape, leading to formation of spatially ordered and highly spectrally uniform QDs. What’s more, the presence of sufficiently small nanoscale mesa owing to the presence of mesa free sidewalls leads to substantial strain relaxation which enables the accommodation of QD forming material combinations with significant lattice mismatch using SESRE, unlike growth in arrays of pits that restrict the material combination to nearly lattice matched (25).
**Fig. S1. SESRE approach for the formation of MTSQD.** (a) Schematic of the MTSQD (red region) forming at apex of each mesa whose spatial selective formation is realized due to the directed atom migration from side of mesa to top (marked by black arrowed) driven by surface-curvature induced stress gradient. (b) Cross-section TEM image of the size-reducing growth on mesa tops with GaAs (dark) and AlGaAs (light, marker layer) [this figure is taken from Ref. (69)]

Following SESRE approach, MTSQDs with \{103\} side walls have been grown and studied (17, 27). As-grown non-planarized 5×8 array of 4.25ML In_{0.5}Ga_{0.5}As MTSQDs (Fig. S2(a)) show spectral non-uniformity of ~6-9nm (Fig. S2(b)) centered around 920-930nm from our previous studies (17, 27). The planarized 5×8 array of 4.25ML In_{0.5}Ga_{0.5}As MTSQDs (Fig. S2(c)) shows a spectral uniformity of 2.8nm (Fig. S2(d), same as Fig. 1(b)) with a blue shifted center wavelength around 890nm. The thermal annealing during the planarization growth induces intermixing of In and Ga in the MTSQDs that shifts the wavelength and reduces the nonuniformity. The observed spectral uniformity is largely limited by the alloy fluctuation. By reducing the alloy fluctuation, we find that the as-grown non-planarized 5×8 array of binary InAs MTSQDs have an unprecedented uniformity of 1.8nm (Fig. S2(e)) with clusters of 6-7 QDs emitting within 250μeV (27). Thus, one expects to have spectral uniformity significantly less than 1.8nm for planarized InAs MTSQDs. All the data present here are from as-grown sample without any growth optimization. We expect that MTSQDs have the potential to be of sub-nm scale spectral nonuniformity.
2. Optical Setup for Resonant measurement

All single photon emission characteristics data reported in the manuscript has been measured using resonant excitation scheme. With the sample containing the MTSQDs mounted in the cryostat, the measurements are done in vertical excitation and vertical detection geometry as shown in Fig. S3. The MTSQDs are excited resonantly at their neutral exciton emission wavelengths using a Ti:Sa Mode lock laser with pulses of 3ps width. The scattered laser light is filtered out using a cross-polarization configuration (using two polarizers and one polarizing beam splitter) as shown in Fig. S3. The exciting laser electric field is along the [110] direction with an accuracy of ±3° and the photons with polarization along [1 1 0] direction are detected, also with accuracy of ±3°. A cross-polarized extinction ratio >1x10^7 is established for the resonant excitation studies reported here. The collected photons from the MTSQDs are spectrally resolved with a spectrometer with 15μeV resolution to reveal the true emission linewidth from the MTSQD and also to act as high-resolution spectral filter to filter out unwanted scattered laser light. The collected photons are then detected by superconducting nanowire detectors.
Fig. S3. Measurement Instrumentation. Schematic of the measurement instrumentation including the Hong-Ou-Mandel interferometry for resonant excitation where the scattered excitation laser light is filtered out with a cross-polarization configuration.

For the study of indistinguishability, we use the Ti-Sa laser to generate pairs of excitation pulses of width ~3ps with a time separation $\Delta t=2$ns, controlled by an unbalanced Michelson interferometer built on the laser side. The impinging laser is also polarized along [110]. The emitted photons from the MTSQD are passed through the polarizer to select emission polarized along $[\bar{1}10]$ and are fed through the spectrometer to the first 50/50 beam splitter of the Hong-Ou-Mandel interferometer (Fig. S3). Photons passing through the beamsplitter are coupled to polarization maintaining fibers through two collimators at the transmitted and reflected ports of the beamsplitter. The path length difference is designed to match the time delay (2ns) of the excitation pulse. Photons passed through the fibers interfere at the fiber-inline-beam splitter (50/50) and detected by the supercomputing nanowire detectors. A $\lambda/2$-waveplate allows for rotating the polarization of photons in one arm of the interferometer with respect to the other arm to make the photons distinguishable on purpose for reference measurement. The instrument response function for the HOM measurement is ~50-100ps.

3. Photon collection efficiency and MTSQD quantum efficiency.

To enhance the collection efficiency of photons emitted from the MTSQDs, we have grown a planarized 5×8 array of 4.25ML In$_{0.5}$Ga$_{0.5}$As MTSQD on DBR (schematic shown in Fig. 1(a)). The cross-section of the as-grown sample is shown in Fig. S4 (a) with the QD a marked as red dot. The typical PL spectrum from one of the MTSQDs sitting on the DBR is shown in Fig. S4(b) measured under above-gap excitation condition (50% of saturation power, with 640nm excitation) at 19.5K. To evaluate the enhancement of photon collection efficiency coming from bottom DBR mirror, we have grown a 4.25ML In$_{0.5}$Ga$_{0.5}$As MTSQD sample sitting on GaAs without DBR as comparison. We have measured PL from MTSQD on a DBR (Fig. S4(b)) and MTSQD on GaAs substrate (Fig. S4(c)) as comparative measurement to establish the enhancement of collection.
efficiency with DBR mirror. PL from a MTSQD without DBR (panel c, red curve) and a MTSQD with DBR (panel b) underneath are measured with the same optical setup and under same normalized power (50% of saturation power, with 640nm excitation) at 19.5K. The photon counts at detector is improved by a factor of ~10. The designed DBR has a reflectivity >95% in the range of 880-930nm. The sample structure (Fig. S4(a)) is designed to enhance the photon collection efficiency at the first objective lens by ~12%. Fig. S4 (d) shows finite element method based simulation of collection efficiency at the first objective lens. The measured PL signal enhancement is consistent with the simulated results, suggesting a factor of 10 enhancement in collection efficiency (the ratio of \( \frac{\text{# of photons collected by first objective}}{\text{# of photons emitted from MTSQD}} \)).

**Fig. S4. Optical signal enhancement with DBR.** PL from a typical MTSQD sitting on DBR (panel (b), SEM of sample in panel (a)) and a typical MTSQD sitting on the GaAs (panel (c)) measured with 640nm excitation at 50% of saturation power at 19.5K. The finite element method simulation of collection efficiency at the first objective lens- indicating 10× enhancement by the DBR is shown in panel (d).

To quantitatively access the quantum efficiency of the as-grown MTSQD (Fig. 1(a) and Fig. S4(a)), we have measured the PL from the MTSQDs under resonant excitation and studied its emission as a function of power under resonant excitation. Fig. S5 shows the PL from one typical MTSQDs and the power dependence of the emission as a function of excitation power. At \( \pi \) pulse, one exciton is created inside the MTSQD per pulse. The detected photon count is 17K/sec. For estimation of the internal quantum efficiency, we calibrated the detection efficiency of the setup (shown in Fig. S3). We have used the laser tuned to the QD emission wavelength (~890nm) to calibrate the detection efficiency of the setup (the ratio of \( \frac{\text{Detection counts at detector}}{\text{# of photons collected by the first objective}} \)).

The detection efficiency of the setup (Fig. S3) is found to be \( \sim (1.81\pm0.02)\times10^{-3} \). Given the photon counts at detector being \( \sim 17K/sec \) at \( \pi \) pulse (Fig. 3(b)), the total number of photon collected by the first objective is \( \sim 0.94\times10^7/sec \). Accounting for the 78MHz repetition rate of the laser and the 12% collection efficiency of emitted photons, we estimate internal quantum efficiency \( \sim 100\% \).
Fig. S5. Resonant PL and Rabi Oscillation. (a) Photoluminescence emission spectra from one typical MTSQD excited under resonant excitation with π/2 pulse, 19.6nW (1.6W/cm²) at 19.5K. The red curve is a Lorentzian fit showing a measured linewidth of 30μeV. (b) Power dependent behavior of peak intensity vs. the square root of laser power (proportional to excitation pulse area) showing clear Rabi oscillation. Error of the measured intensity is within the symbol size. The inserted Bloch sphere represents the switching from ground state |0⟩ (empty QD) to one exciton state |1⟩ (Fig. 3(b), recaptured here for easy reference).

4. Single Photon Purity.

To assess the general behavior of single photon emission purity of the MTSQDs under resonant excitation, we have shown in Fig. 3(a) measured second-order correlation function of the photon emission from one MTSQD (2,2) (marked by the row and column number) in the array at 19.5K. Here we show results from one other MTSQD (MTSQD (4,2) in the array) in Figure S6 below. By calculating the ratio of the area under the τ=0 peak and the average of areas under the τ≠0 peaks, we obtain a $g^{(2)}(0)$ of 0.05 for the MTSQD, giving single photon emission purity ~97.5%. The measured single photon purity is consistent with previously reported behavior of MTSQDs (17, 27). A calculated curve based on theory (29) is shown as the red line in Fig. 3(a) and Fig. S6. The number of photons emitted from QD (n) follows $n(t) \sim \exp \left(-\frac{t}{T_1}\right)$ where $T_1$ is the PL decay lifetime of the QDs. Thus, the second order correlation function $g^{(2)}(\tau)$ is of the form (around each peak),

$$g^{(2)}(\tau) = \langle n(t)n(t+\tau) \rangle \sim \exp \left(-|\tau - t_{exc}|/T_1\right).$$

The coincidence count histogram is of the form

$$h(t) = A \int IRF(t)[\sum_{m=0} \exp \left(-|\tau - t - mT_1|/T_1\right) + g^{(2)}(0) \exp \left(-|\tau - t_1|/T_1\right)]dt,$$

where $IRF(t)$ is the instrument response function, $T$ is the time interval between excitation pulses, and $m$ is an integer sequence number for an individual excitation pulse. The $g^{(2)}(0)$ extracted from theory is consistent with that extracted based on peak area ratios.
Fig. S6. Single photon emission purity. Histogram of coincidence counts of emission from MTSQD (4,2) using HBT setup with resonant excitation and with π/2 pulse at 19.5K. The red curve is the calculated curve based on theory (29).

5. Decay Dynamics:

MTSQD (2, 2) (shown in Fig. 1 and 2 of manuscript) provides single photons in coherent superposition of two finely split state with ∆~6.4μeV (as shown in the beating signal in time-resolved PL of Fig. 3(c)). The observed beating pattern in time-resolved PL is also observed in other MTSQDs. Fig. S7 shows the measured TRPL data on MTSQD (4,2) at 19.5K under resonant excitation with π/2 pulse. It also shows beating pattern and reveals ∆ = 3.8μeV and $T_1^{(a)} = T_1^{(b)} = 0.55\text{ns}$ from fitting (red line, Fig. S7) using Eq. 1. These data suggests that (1) MTSQDs have fine structure splitting <10ueV and (2) the emitted single photons are in coherent superposition of the states from the two finely split states.

Fig. S7. Time-resolved PL. Measured time-resolved fluorescence from the MTSQD (4,2) excited under resonant excitation with π/2 pulse at 19.5K. The red curve is fitting to the data using Eq. (1) from our three-level model.
6. HOM Visibility in the MTSQD array:

Our study of the HOM two-photon interference behavior has been limited to three randomly chosen MTSQDs at a temperature ~20K and one at ~12K. This is owing to our cryogen-free cryostat cold-head having degraded over time and limiting sample temperature to ~21/22K. Turning over to an older standard liquid He continuous flow cryostat has, regrettably, not been an option owing to the prohibitive cost ($85/liter for us) of LHe. Nevertheless, as having a HOM measurement on at least one MTSQD at the low temperatures (customarily near or below 4K in the literature) is essential, we obtained enough LHe to be able to carry out measurements on one MTSQD. Regrettably, the cryostat did not cool below 11.5K and so we gathered all optical data, including HOM, at this temperature on one MTSQD. Thus, Fig. 5 of the main text shows the HOM visibility for this one MTSQD at two temperatures.

Here we provide added HOM information at ~22K from the other two MTSQDs. We find the measured visibility around ~20K to 22K from all three MTSQDs, after correction of multiphoton emission events, are 0.57, 0.56 and 0.49 yielding an average value of V~ 0.54 with a standard deviation of 0.04.

![Figure S8. Photon Indistinguishability.](image)

(a) A representative two photon interference coincidence count histogram of one of the two additional MTSQDs studied in addition to MTSQD (2,2) reported in the main text. A visibility of ~0.56±0.1 is obtained (corrected for multiphoton emission events) at 22.6K. (b) Zoomed-in plot around zero-peak for better clarity. We note that HOM measurements on three MTSQDs show an overall visibility ~0.54 ± 0.04 for ~20 to 22K temperature range.

A representative two photon interference (TPI) coincidence count histogram from one of these two MTSQDs is shown in Fig. S8. We note that, the histogram was collected using APD detectors that have a large ~300ps time jitter compared to the high timing resolution (50 to 100ps) superconducting nanowire detectors that was used for the data shown in Fig. 4 and 5 of the main manuscript. The higher detector jitter has thus resulted in the absence of the oscillatory behavior of the individual peaks in the HOM coincident count spectra- but has no effect on the estimated value of the Visibility.

7. Indistinguishability: Effect of Phonon and Spectral Diffusion.

The effect of phonon and spectral diffusion on TPI visibility is analyzed following the model reported in Ref. (4J) using Markovian approximation in addressing the phonon induced dephasing time. The TPI visibility is expressed (4J) as
\[ V_c(T) = \frac{\Gamma}{\Gamma_{SD} + \gamma(T) + \Gamma} \]

where \( \Gamma = \frac{1}{2T_1} \),
\[ \gamma(T) = \frac{\gamma_0}{\exp\left(\frac{\alpha}{T}\right)-1} \left[ \frac{1}{\exp\left(\frac{\alpha}{T}\right)-1} + 1 \right] \] is the phonon induced dephasing rate and \( \Gamma_{SD} \) is the spectral diffusion-induced dephasing rate. We used the reported \( \gamma_0 \) and \( \alpha \) from Ref. (41) to fit to our measured data shown in Fig. 5(a). Given the measured TPI visibility data both at 19.5K and 11.5K, the fitted behavior of visibility as a function of temperature is shown in Fig. S9 (black line in Fig. S9, measured data shown as red dots). The result matches the typical known temperature dependence of the exciton dephasing time in InGaAs/GaAs material system reported in Ref. (41) and indicates an expected visibility \( \sim 92\% \) at 4K.

**Fig. S9. Phonon Effect on photon visibility.** Measured visibility (red squares) and fit (black curve) for temperature dependent visibility without any Purcell enhancement. The red curve and the blue curve indicate expected visibility when a Purcell enhancement \( \sim 5 \) and \( \sim 10 \), respectively, are introduced.

The visibility can be further enhanced by embedding the MTSQD in appropriate photonic cavity structures introducing a Purcell enhancement (7) - thus shortening the radiative lifetime \( T_1 \). With a Purcell enhancement \( F_p \), the expected TPI visibility can be expressed as
\[ V'_c(T) = \frac{\gamma \times F_p}{\Gamma_{SD} + \gamma(T) \times F_p + \Gamma \times F_p} \]

In Figure S9, we show the expected Visibility for a Purcell enhancement \( \sim 5 \) (red curve) and \( \sim 10 \) (blue curve) - typically achieved via DBR micropillar (7), photonic crystal membrane (70), or dielectric nanoantenna (62) structures. At 4K, TPI visibility larger than 97\% is estimated for Purcell enhancement 5, indicating that near unity indistinguishability can be readily reached for the MTSQD SPSs.

**8. Three-level model.**

In this section we provide additional detailed information on the analysis of the photoluminescence, time resolved emission, single photon self-interference, and two-photon
interference (TPI) of photons emitted from the MTSQDs based on the model of three-level structure of the exciton manifold. The content is organized as follows:

- In Section 8.a, we define the MTSQD as a three-level system and define a simple Hamiltonian that captures the resonant excitation and detection system. With this model, we derive (a) the power dependent PL intensity (Rabi oscillation) and (b) the expression of wavepacket of the emitted photon including the beating effect from the two fine structure split (FSS) states to compare with the data observed in time resolved photoluminescence measurement.

- In Section 8.b, exploiting the photon wavepacket derived in section 8.a and adapting the approach of Bylander et. al (71) we show the analysis of outcome of photon self-interference and two photon Hong-Ou-Mandel interference to interpret the experimental measurements presented in this paper.

8.a. Three-level Model of Dynamical Evolution of Exciton in MTSQD SPS:

The measured time resolved photoluminescence of the MTSQDs as shown in Fig. 3(c) of the main text indicates a three-level structure of the ground level exciton of the MTSQD. In this section we analyze resonant fluorescence behavior of such a system shown schematically in Fig. S10(a). The two FSS split exciton states are denoted as |1a⟩ and |1b⟩ with energy separation Δ, and the ground state (no exciton) is denoted as |0⟩. Past studies have shown (30) that the transition dipole moments of these two fine structure split states are linearly polarized at an angle ~20°-30° with respect to the crystallographic direction [110] and [1 1 0]. Here in this analysis we depict this angle as φ0 — as indicated in Fig. S10(b).

With the above assumption, we express the Hamiltonian corresponding to resonant excitation and detection measurement with the excitation E-field polarization along the [1 1 0] (zinc-blende) direction, and emitted photons polarized in the [1 ̅ 1 0] direction being collected into detection optics [Fig. S10(c)]. The overall Hamiltonian is expressed as,

\[ H = H_{MTSQD} + H_{Laser-MTSQD} + H_{Cav} + H_{QD-Cav} + H_{Cav-Det} \] (1)

Here, the first term, Hamiltonian of the exciton manifold of the MTSQD by itself is,

\[ H_{MTSQD} = \left( \omega_a - \frac{i}{2T_1^{(a)}} + F(t) \right) \sigma_a^+ \sigma_a + \left( \omega_b - \frac{i}{2T_1^{(b)}} + F(t) \right) \sigma_b^+ \sigma_b \] (2)

in which \( \omega_a \) and \( \omega_b \) represent the energy of the exciton states |1a⟩ and |1b⟩, respectively, and the effect of the radiative decay is captured as the introduced non-Hermiticity through decay times, \( T_1^{(a)} \) and \( T_1^{(b)} \). For simplicity, \( \hbar \) is normalized to unity. \( F(t) \) is the random energy shift fluctuating with time \( t \) introduced by phonons and spectral diffusion, and is modelled by Langevin approach with Markovian approximation assuming a memoryless thermal reservoir (71). Thus we have \( \langle F(t_1)F(t_2) \rangle = \frac{2}{T_2^*} \delta(t_1 - t_2) \), where \( T_2^* \) is the dephasing time. Here \( \langle \cdot \rangle \) denotes statistical average.
Fig. S10. Three-level model of MTSQD. (a) Schematic of the three level structure of the MTSQD. (b) The orientations of the transition dipoles of the two exciton states with respect to the [110] type direction taken as an angle $\phi_0$. (c) The measurement geometry indicating the different components of the Hamiltonian as shown in equation (1).

We use the $\vec{p} \cdot \vec{E}$ type interaction to express in equation (1) the interaction term $H_{\text{Laser-MTSQD}}$ between the excitation laser and the exciton states. Under rotating wave approximation,

$$H_{\text{Laser-\text{MTSQD}}} = p_0 E_0 \ e^{-\frac{2(t-t_0)^2}{\Delta t^2_{\text{Laser}}}} \ [\cos \phi_0 \ \sigma^+_a e^{-i\omega_{\text{Exc}} t} + \sin \phi_0 \ \sigma^+_b e^{-i\omega_{\text{Exc}} t} + h.c.]$$

Here $E_0$ is the peak E-field strength (polarized along the [1 1 0] direction) of the excitation laser. $\Delta t_{\text{Laser}}$ is the laser pulse width, $\sim$3ps for our case, and $\omega_{\text{Exc}}$ is the center wavelength of the pulsed laser. The transition dipole moments of the two exciton states are assumed to have equal amplitude $p_0$.

The emitted photons from the MTSQD are coupled to the cavity formed by the planar DBR bottom mirror and the GaAs-Air interface at the top. Photons from this DBR cavity are quickly leaked into the detection optics mode. This photon collection process is captured using the last three terms of the Hamiltonian in equation (1) as,

$$H_{\text{Cav}} = (\omega_{\text{Cav}} - i \kappa_{\text{Cav}}) \ a^+_{\text{Cav}} a_{\text{Cav}}$$

$$H_{\text{QD-Cav}} = g \ [\sin \phi_0 \sigma^+_a e^{-i\omega_{\text{Cav}} t} - \cos \phi_0 \sigma^+_b e^{-i\omega_{\text{Cav}} t} + h.c.]$$

$$H_{\text{Cav-Det}} = \kappa_{\text{Cav}} a^+_{\text{Det}} a_{\text{Cav}}$$

Here $\kappa_{\text{Cav}}$ is the rate of photon leakage from the DBR-GaAs cavity to the detection optics. Transforming to the rotating frame given by $H_0 = \omega_a \sigma^+_a \sigma_a + \omega_b \sigma^+_b \sigma_b + \omega_{\text{Cav}} a^+_{\text{Cav}} a_{\text{Cav}}$, the Hamiltonian (sum of (2) to (6)) is simplified to $H = H_0 + e^{-iH_0 t} \ H_I \ e^{iH_0 t}$, where,
\[
H_1(t) = p_0 E_0 e^{- \frac{(t-t_0)^2}{2 \Delta t_{\text{Laser}}^2}} \left[ \cos \phi_0 \sigma_a^+ e^{i \omega_a - i \omega_{\text{Exc}} t} + \sin \phi_0 \sigma_b^+ e^{i \omega_b - i \omega_{\text{Exc}} t} + h. c. \right] \\
+ g \left[ \sin \phi_0 \sigma_a^+ e^{i \omega_a - i \omega_{\text{Cav}} t} - \cos \phi_0 \sigma_b^+ e^{i \omega_b - i \omega_{\text{Cav}} t} + h. c. \right] \\
+ \kappa_{\text{Cav}} a^\dagger_{\text{Det}} a_{\text{Cav}} e^{- i \omega_{\text{Cav}} t} + F(t) \sigma_a^+ \sigma_a + F(t) \sigma_b^+ \sigma_b - i \kappa_{\text{Cav}} a^\dagger_{\text{Cav}} a_{\text{Cav}} - \frac{i}{2 \tau_1(a)} \sigma_a^+ \sigma_a - \frac{i}{2 \tau_1(b)} \sigma_b^+ \sigma_b \quad (7)
\]

The evolution of the combined state of the MTSQD exciton and the emitted photon under resonant excitation therefore follows,

\[
i \frac{d}{dt} |\Psi_1(t)\rangle = H_1(t) |\Psi_1(t)\rangle \quad (8)
\]

with the state of the system, in the rotating frame, expressed as,

\[
|\Psi_1(t)\rangle = c_0(t) |0\rangle + c_a(t) \sigma_a^+ |0\rangle + c_b(t) \sigma_b^+ |0\rangle + c_{\text{Cav}}(t) a^\dagger_{\text{Cav}} |0\rangle + c_{\text{Det}}(t) a^\dagger_{\text{Det}} |0\rangle \quad (9)
\]

Equation (8) and (9) provide us the basic framework to analyze the resonant excitation measurements of PL from the MTSQD modelled as the three-level system. Specifically, this framework is now used to analyze the power-dependent and time-dependent PL response as follows:

**Power Dependent Resonant PL - Rabi Oscillation**

In the PL measurements, the QD exciton is first initialized by a short (~3ps) excitation laser pulse. Let us denote time \( t = 0 \) as the time at which the exciton state has just been initialized. Using the Hamiltonian in eq. (7), and under the assumption that the excitation pulse is much shorter (~3ps) compared to the decay timescale (~350ps), we get the state of the system at the end of the excitation pulse to be,

\[
|\Psi_1(0)\rangle = \sin \left( \int_{\text{Pulse}} \Omega(t) dt \right) [\cos \phi_0 \sigma_a^+ + \sin \phi_0 \sigma_b^+] |0\rangle \quad (10)
\]

where \( \Omega(t) = p_0 E_0 e^{- \frac{(t-t_0)^2}{2 \Delta t_{\text{Laser}}^2}} \). Note that both the states \(|1_a\rangle\) and \(|1_b\rangle\) are populated by the laser since the excitation E-field has components along the transition dipole moment of both these exciton states (Fig. S10). For a Gaussian laser pulse of repetition rate \( F_{\text{Laser}} \), full width half maximum is \( \Delta t_{\text{Laser}} \), laser spot area = \( A_{\text{Spot}} \), and the average incident power on the GaAs surface \( P_{\text{inc}} \), we estimate,

\[
\int_{\text{Pulse}} \Omega(t) dt = p_0 \sqrt{\frac{P_{\text{inc}}}{F_{\text{Laser}} A_{\text{Spot}}}} \frac{T_{\text{GaAs}} \eta_{\text{GaAs}} 2 \sqrt{\pi}}{\ln 2} \frac{\Delta t_{\text{Laser}}}{\Delta t_{\text{Laser}}} = C p_0 \sqrt{P_{\text{inc}}} \quad (11)
\]

Here \( F_{\text{Laser}} \) is the laser pulse rate ~78 MHz for us; \( \eta_{\text{GaAs}} \) is electromagnetic impedance of GaAs ~ 110 Ohm; \( T_{\text{GaAs}} \) is the transmittance of the air-GaAs interface ~ 66%. \( A_{\text{Spot}} \) is the laser spot area~1.22 \( \mu \)m\(^2\). The effect of these constants is combined into \( C = \frac{T_{\text{GaAs}} \eta_{\text{GaAs}} 2 \sqrt{\pi}}{\sqrt{F_{\text{Laser}} A_{\text{Spot}} \ln 2} \Delta t_{\text{Laser}}} \), which is a constant that only depends on the measurement instrumentation. \( P_{\text{inc}} \) is the average excitation power impinging on the sample surface. Equation (10) and (11) indicate the Rabi oscillation of the amplitude of the exciton states with respect to \( P_{\text{inc}} \) via the proportionality,

\[
\text{Intensity} \ \propto \ \sin^2 \left( C p_0 \sqrt{P_{\text{inc}}} \right) \quad (12)
\]

This oscillatory behavior manifests itself in the power dependent PL intensity data shown in Fig. 3(b). Note, in this analysis we did not take into account the excitation induced dephasing process.
(72) – which results in the decay in the oscillatory amplitude of the power dependent PL as seen in Fig. 3(b) of the main text. Nevertheless, comparing the measured oscillatory period of the power-dependent PL emission with equation (12), we estimate that the transition dipole moment of the exciton states to be of magnitude $p_o \approx 70$ Debye.

**Time-Dependent Resonant PL: Description of Emitted Photon Wave Packet**

Once the excitons in states $|1_a\rangle$ and $|1_b\rangle$ are populated by the excitation laser pulse to the state $|\Psi_1(0)\rangle$ given in equation (10), the process of single photon emission is analyzed using the same Hamiltonian as in equation (7). Under the expansion shown in equation (9), applying the Hamiltonian in equation (7) and Schrödinger equation in eq. (8), we have,

\[
i \frac{d}{dt} c_a(t) = \left[ -\frac{i}{2T_1^{(a)}} + F(t) \right] c_a(t) + g \sin \phi_0 \ e^{-i\omega_{cav}t-i\omega a t} \ c_{cav}(t)
\]

\[
i \frac{d}{dt} c_b(t) = \left[ -\frac{i}{2T_1^{(b)}} + F(t) \right] c_b(t) - g \cos \phi_0 \ e^{-i\omega_{cav}t+i\omega b t} \ c_{cav}(t)
\]

\[
i \frac{d}{dt} c_{cav}(t) = -i\kappa_{cav} c_{cav}(t) + g \sin \phi_0 \ e^{i\omega_{cav}t-i\omega a t} \ c_a(t) - g \cos \phi_0 \ e^{i\omega_{cav}t-i\omega b t} \ c_b(t)
\]

and,

\[
i \frac{d}{dt} c_{Det}(t) = \kappa_{cav} e^{-i\omega_{cav}t} c_{cav}(t)
\]

The evolution can be further simplified by applying the weak coupling limit. The QD-cavity coupling strength $g$ in our planar DBR cavity is $\sim 1 \mu eV$, much smaller than the decay rate $\kappa_{cav} \approx 10 \text{meV}$. Thus, $\kappa_{cav} \gg g$ which leads to $|c_{cav}(t)| \ll |c_a(t)|, |c_b(t)|$. Under this weak coupling limit, solving equations (13) to (16), we have

\[
c_a(t) \approx e^{-\frac{t}{2T_1^{(a)}}} e^{-i \int_0^t F(t) \, dt} \ c_a(0)
\]

\[
c_b(t) \approx e^{-\frac{t}{2T_1^{(b)}}} e^{-i \int_0^t F(t) \, dt} \ c_b(0)
\]

The integrated effect of the random energy fluctuation $F(t)$ is reflected as a randomized additional phase $\phi(t) = \int_0^t F(t) \, dt$ that represents the dephasing process. From Langevin formulation (71) based on a memoryless reservoir we have

\[
\langle e^{-i\phi(t_1)} e^{i\phi(t_2)} \rangle = e^{-\frac{|t_1-t_2|}{\tau_2}}
\]

Finally, the amplitude of the photon state collected in the detection optics mode can be expressed, from equation (16) to (17), as,

\[
c_{Det}(t) = g \int_0^t e^{-i\omega_{cav}t} \left[ \sin \phi_0 \ e^{-i\omega a t} \ c_a(t) - \cos \phi_0 \ e^{-i\omega b t} \ c_b(t) \right] \, dt
\]

Using equation (18) and (19), respectively, as the expressions for $c_a(t)$ and $c_b(t)$, and using equation (10) for the state at $t = 0$, we get the emitted photon wavepacket collected in the detection optics expressed as,
\[ |\Psi_{\text{photon}}(t)\rangle = \int_{0}^{t} f(t) a_{\text{Det}}^\dagger(t) |0\rangle \, dt \]  

(22)

Where \( f(t) = g \sin \phi_0 \cos \phi_0 \sin(f_{\text{Pulse}} \Omega(t) dt) \left[ e^{-i \omega_a t - \frac{t}{2T_1^{(a)}}} - e^{-i \omega_b t - \frac{t}{2T_1^{(b)}}} \right] e^{-i \phi(t)}. \)

equation (22), we can express the time resolved intensity as,

\[ I(t) \propto |f(t)|^2 \propto \left| e^{-i \Delta t} e^{-\frac{t}{2T_1^{(a)}}} - e^{-\frac{t}{2T_1^{(b)}}} \right|^2 \]  

(23)

This analytical form is shown in equation (1) of the main text and is used to fit the measured time resolved PL data as shown in Fig. 3(c) of the main text and Fig. S7 to extract \( \Delta, T_1^{(a)}, \) and \( T_1^{(b)}. \)

8.6 Photon Interference Measurements:

The photon wavepacket expression derived in last section is now exploited to derive the expected outcome of single photon self-interference and two-photon interference, as follows:

**Self-Interference of the emitted photon - Fringe Contrast:**

Self-interference is exploited in the coherence time measurement, where the photon wavepacket emitted into the detection optics is split into two branches and then recombined into one branch with a relative delay of \( \tau_D \), as indicated in Fig. S11.

\[ |\Psi_{\text{photon}}\rangle = \int_{0}^{\infty} dt' f(t') a^\dagger(t') |0\rangle \]

Fig. S11. Photon self-interference diagram. Schematic of photon correlation measurement set-up to determine self-interference of a single photon.

Using the photon wavepacket expression in equation (22), the resultant wavepacket at the detector can be expressed as,

\[ |\Psi_{\text{at detector}}(t)\rangle \propto \int_{0}^{t} f(t) a_{\text{Det}}^\dagger(t) |0\rangle \, dt + \int_{0}^{t} f(t + \tau_D) a_{\text{Det}}^\dagger(t) |0\rangle \, dt \]  

(24)

Thus, intensity at the detector can be expressed in the following proportionality relation:

\[ I_{\text{Det}} \propto \int_{0}^{\infty} |f(t)|^2 \, dt + \text{Re} \left[ \int_{0}^{\infty} f(t) f^*(t + \tau_d) \, dt \right] \]  

(25)

The fringe contrast is expressed as,

\[ \text{Fringe contrast} = \frac{\max(I_{\text{Det}}) - \min(I_{\text{Det}})}{\max(I_{\text{Det}}) + \min(I_{\text{Det}})} = \frac{\int_{0}^{\infty} f(t) f^*(t + \tau_d) \, dt}{\int_{0}^{\infty} |f(t)|^2 \, dt} \]  

(26)
\[
\frac{1}{I_0} \left[ \int_0^\infty dt \ e^{-\frac{t}{\Delta}} \sin \left( \frac{\Delta}{2} t \right) \sin \left( \frac{\Delta}{2} (t + \tau_d) \right) \right] e^{-\frac{\tau_d}{2T_1}} \left( e^{-i\phi(t) + i\phi(t+\tau_d)} \right) = 1 \int_0^\infty dt \ e^{-\frac{t}{\Delta}} \sin \left( \frac{\Delta}{2} t \right) \sin \left( \frac{\Delta}{2} (t + \tau_d) \right) \right] e^{-\frac{\tau_d}{2T_1}} - \frac{\tau_d}{T_2} (27)
\]

Here \( I_0 = \int_0^\infty |f(t)|^2 dt = \frac{\Delta^2 \tau_d^2}{2(1 + \Delta^2 \tau_d^2)} \) is the intensity of a single photon wavepacket. Equation (28) provides the analytical expression for the fringe contrast that is shown in equation (2) of the main text. This equation is used to fit the data in Fig. 3(d).

Two Photon Interference- Hong-Ou Mandel Configuration:

In the two photon HOM interference measurement, two different photon wavepackets emitted from the same MTSQD with a time difference of \( \tau_d \approx 2\text{ns} \) are collected into the detection optical branch. One of these two photons is delayed, also by \( \tau_d \) to compensate for the emission time difference and then interfered with the other photon. Schematically this is shown in Fig. S12.

![Two photon interference path diagram](image)

**Fig. S12. Two photon interference path-diagram.** A schematic representation of the HOM two-photon interference measurement.

We adapt the approach of Bylander (71) and apply it for the single photon wave packet shown in equation (22) to derive the analytical expression of the two-photon correlation function as,

\[
g^{(2)}(t_1,t_2) = |f(t_1 - \tau_d) f(t_2 - 2\tau_d)|^2 + |f(t_2 - \tau_d) f(t_1 - 2\tau_d)|^2 + |f(t_1) f(t_2 - 2\tau_d)|^2 + |f(t_2) f(t_1 - 2\tau_d)|^2 + |f(t_1 - \tau_d) f(t_2 - \tau_d)|^2 [2 - \left( e^{-i\phi_1(t_1) + i\phi_1(t_2) - i\phi_2(t_2) + i\phi_2(t_1)} \right) + c.c.] \]

(29)

The photon interference effect is contained in the time average of the random phase fluctuation, \( \phi_1(t) \) and \( \phi_2(t) \), of the first and the second photon emitted from the same QD, separated by \( \approx 2\text{ns} \). Since 2ns is much longer than the dephasing timescale at the operating temperature of 19.5K, it is reasonable to assume that \( \phi_1(t) \) and \( \phi_2(t) \) are uncorrelated (71). Thus,

\[
\langle e^{-i\phi_1(t_1) + i\phi_1(t_2) - i\phi_2(t_2) + i\phi_2(t_1)} \rangle = \left( e^{-i\phi_1(t_1) + i\phi_1(t_2)} \right) \left( e^{-i\phi_2(t_2) + i\phi_2(t_1)} \right) = e^{-\frac{2|t_1 - t_2|}{T_2}} = e^{-\frac{2|\tau|}{T_2}} (30)
\]

The time resolved HOM \( g^{(2)}(t_1,t_2) \) as shown in equation (29) is plotted in Fig. 3(c) of the main text along with the measured result. Further, from equation (29) and (30), integrating over \( t_2 \)- the detection time of the “stop” detector, the analytical expression of the HOM \( g^{(2)}(\tau) \) coincidence count histogram can be derived as,
\begin{equation}
g_{\parallel}^{(2)}(\tau) = \int_{0}^{\infty} dt \, e^{-\frac{2t}{T_1}} \sin^2 \left( \frac{\Delta}{2} t \right) \sin^2 \left( \frac{\Delta}{2} (t + |\tau|) \right) \left[ 1 - e^{-\frac{2|\tau|}{T_2}} \right] e^{-\frac{|\tau|}{T_1}} \tag{31}
\end{equation}

For perpendicular polarization, i.e., no interference at the second HOM beam splitter, the center peak of the HOM coincidence count takes the form,

\begin{equation}
g_{\perp}^{(2)}(\tau) = \int_{0}^{\infty} dt \, e^{-\frac{2t}{T_1}} \sin^2 \left( \frac{\Delta}{2} t \right) \sin^2 \left( \frac{\Delta}{2} (t + |\tau|) \right) \quad e^{-\frac{|\tau|}{T_1}} \tag{32}
\end{equation}

These functional forms in equation (31) and (32) are used to fit to the measured HOM $g^{(2)}$ data shown in Fig. 3 of the main text and to extract the dephasing time $T_2^*$ of the exciton state of the MTSQDs.
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