

Supplementary Information:

This document contains supplementary text discussing the methods used, figures providing information on the QD sample and level structure (Fig. S1), key components of the experimental setup (Fig. S2), experimental verification of the spectral transmission window for the Fabry-Perot cavity used in these experiments (Fig. S3), and a discussion section on optical transition linewidths measured using different techniques (Figs. S4 and S5).

Sample

The InAs/GaAs quantum dots studied were grown by molecular beam epitaxy (MBE) using the partially covered island technique and are embedded in a Schottky diode heterostructure. An illustration of the sample structure is in Fig. S1a. This study is conducted in the single excess electron regime, i.e. at the centre of the single electron charging plateau. Thus, the two spin states of the electron form the ground state manifold which can be coupled via a resonant optical field to the excited state manifold comprising two electrons (forming a spin singlet) and a hole; the X^{-1} transitions. The excited state lifetime leads to a spontaneous emission rate of γ_{sp} , as indicated in Fig. S1b. Due to their particular spin configurations both state manifolds are doubly degenerate and a finite magnetic field is generally used to lift this degeneracy via the Zeeman Effect.

Techniques

A diagram of the experimental setup can be found in Fig. S2. For the measurements the gated QD sample is housed in a magneto-optical bath cryostat and cooled to 4.2 K. A cubic zirconia solid immersion lens (SIL) is mounted on the epitaxial sample surface in order to improve both the light focusing and light gathering power of the fiber-based confocal microscope. We first identify an X^{-1} transition from voltage dependent photoluminescence with an excitation wavelength of 780 nm (PL setup in Fig. S2). Next, a scanable single mode diode laser with 1.2-MHz frequency and 0.5% power stabilization is tuned across the X^{-1} transition (DT setup in Fig. S2) to identify the X^{-1} resonances. The same laser is fixed to the desired detuning from the selected X^{-1} resonance. The light resonantly scattered by the QD is collected through the second arm of fiber confocal microscope, sent through a ~ 34.5 MHz frequency stabilized Fabry-Perot cavity and subsequently analyzed with a liquid nitrogen cooled CCD (RF setup in Fig. S2). The Fabry-Perot normalized transmission is presented in Fig. S3. The measured Fabry-Perot throughput at peak transmission frequency is 30%. To suppress the background laser light and collect the resonance fluorescence spectrum we operate the microscope in a dark-field configuration by placing a linear polarizer in the microscope collection arm perpendicular to the incoming linearly polarized laser field. In this configuration we measure an extinction of the laser light greater than 5×10^3 . In order to obtain the excited state lifetime, Hanbury-Brown and Twiss type photon-correlation measurements are performed by two single photon counting avalanche photodiodes and a record of coincidence events is kept to build up a time-delay histogram ($g^{(2)}$ setup in Fig.

S2). The results under both above bandgap and resonant excitation are presented in Fig. S4. For panel b of Fig. S4 there is no background subtraction and the entire fluorescence triplet (including residual laser background) is sent to the $g^{(2)}$ setup.

A Discussion on Transition Linewidth:

This is a short supplementary note on the transition linewidth measured using conventional nonresonant photoluminescence (PL), differential transmission (DT), resonance fluorescence (RF) and photon correlation ($g^{(2)}$) techniques. In all linewidth discussions in this manuscript we use the below definitions. All spectral measurements result in Lorentzian lineshapes and the intensity correlation measurement results in a double exponential profile.

The functional form of a Lorentzian is:

$$\propto \frac{1}{\Delta^2 + (\gamma/2)^2}$$

where Δ is the independent variable such as laser-transition detuning in DT or emission frequency in PL and RF, and γ determines the transition linewidth. The measured full width at half maximum (FWHM) of this Lorentzian is γ . For optical transitions in which the only dissipation and/or dephasing mechanism is spontaneous emission, which we assume throughout this discussion, γ is equal to the spontaneous emission rate γ_{sp} [1, 2]. In PL the emission line shape L is [1]

$$L(\omega_{em}) \propto \frac{1}{\Delta^2 + (\gamma_{sp}/2)^2}$$

where Δ is the detuning between the emitted light frequency ω_{em} and the transition resonance frequency. We highlight in PL the emission linewidth is exactly equal to γ_{sp} .

1) DT: In DT the extinction line shape is measured directly and should equal [3]

$$\Delta T(\omega_{sc}) \propto 1 - \frac{1}{\Delta^2 + (\gamma_{sp}/2)^2}.$$

The important point here is that the linewidth is now equal to γ_{sp} .

2) RF sidebands: In RF, in the strong excitation laser limit, the side band lineshape is [2]

$$L(\omega_{em}) \propto \frac{1}{(\Delta - \Omega)^2 + (3\gamma_{sp}/4)^2}$$

where Ω is the bare Rabi frequency. In RF the sideband linewidth is equal to $3\gamma_{sp}/2$.

3) RF central peak: Finally, in RF the centerband lineshape, assuming strong excitation, is [2]

$$L(\omega_{em}) \propto \frac{1}{\Delta^2 + (\gamma_{sp}/2)^2}.$$

The linewidth, as in DT, is equal to γ_{sp} . Due to the laser background this peak is inaccessible in the strong excitation regime.

4) $g^{(2)}$ profile: In photon correlation measurements, the correlation lineshape for a single anharmonic optical transition is [2]

$$g^{(2)}(\tau) \propto 1 - \exp(-\gamma_{sp}\tau).$$

This is the well-known anti-bunching dip in the limit of vanishing excitation laser power to eliminate multiple carrier relaxation effects. A fit to the $g^{(2)}$ anti-bunching dip reveals γ_{sp} .

References:

- [1] H. J. Carmichael, *Statistical Methods in Quantum Optics I*, (Springer, 2002).
- [2] R. Loudon, *The Quantum Theory of Light*, 3rd Ed., (Oxford University Press, 2000).
- [3] K. Karrai and R.J. Warburton *Superlattices and Microstructures* **33** 311 (2003).
- [4] M. Schubert, S. I. Siemers, R. Blatt, W. Neuhauser and P. E. Toschek *Physical Review Letters* **68** 3016 (1992).

Supplementary Information- Figure Captions

Figure S1 An illustration of the sample structure and the corresponding energy diagram. A) All experiments discussed here were performed on single QDs isolated in real space and spectrum on the top QD layer (red), with no optical signatures of any other QDs from the lower QD layer (blue). B) The doubly degenerate 2-level system comprised of a single electron ground state and the trion (electron spin singlet and a single hole) excited state in zero magnetic field. The plot presents the simulated spectrum of spontaneous emission from a 2-level system.

Figure S2 A schematic of the experimental setup indicating key elements comprising each technique used in the manuscript.

Figure S3 The measured transmission linewidth of the Fabry-Perot cavity as it is scanned across a 300-KHz linewidth single frequency laser independently stabilized to 1.2 MHz frequency uncertainty over several hours.

Figure S4 A comparison of the raw photon correlation [$g^{(2)}$] measurements. A) Above bandgap excitation at a power well below the saturation level and carrier capture effects displays the true radiative lifetime of the excited state. B) Resonant excitation at moderate power ($\Omega < \gamma_{sp}$) displays the expected antibunching behaviour for the full resonance fluorescence spectrum despite the laser background. The slight bunching behaviour at longer time delays is a consequence of optical pumping and the multi-level structure of the X^{-1} transition [4] and will be discussed elsewhere.

Figure S5 A comparison of the linewidths extracted from A) nonresonant PL transmitted through the same Fabry-Perot cavity used in RF measurements, B) the resonance fluorescence sideband at 1.852 μ W excitation laser power and C) power broadening via DT.

Figure S1:

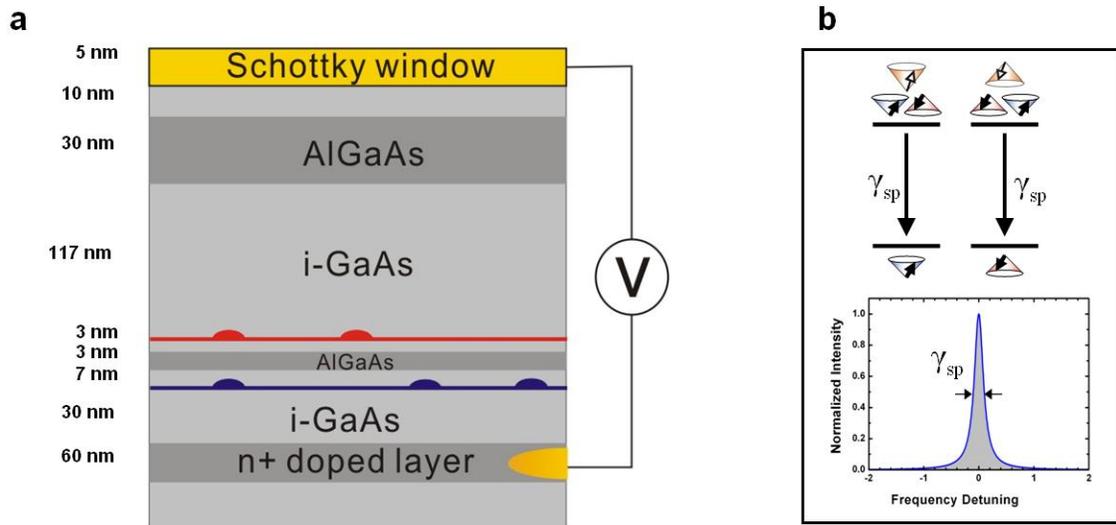


Figure S2:

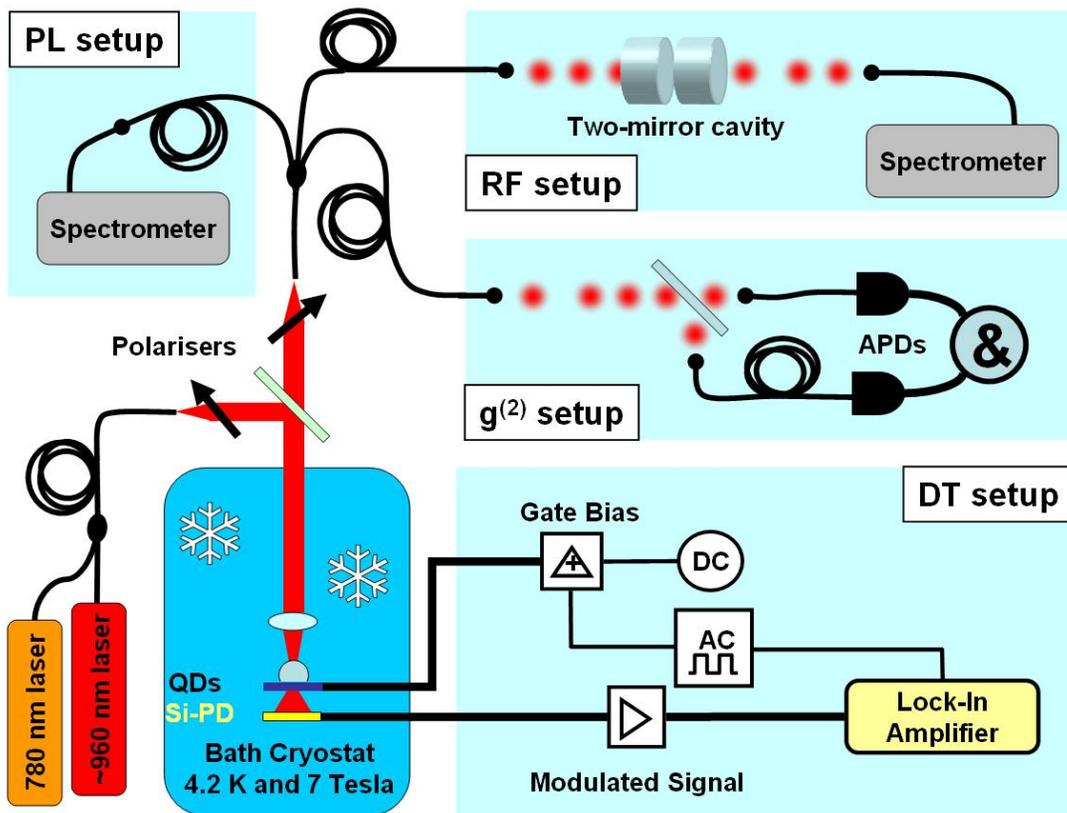


Figure S3:

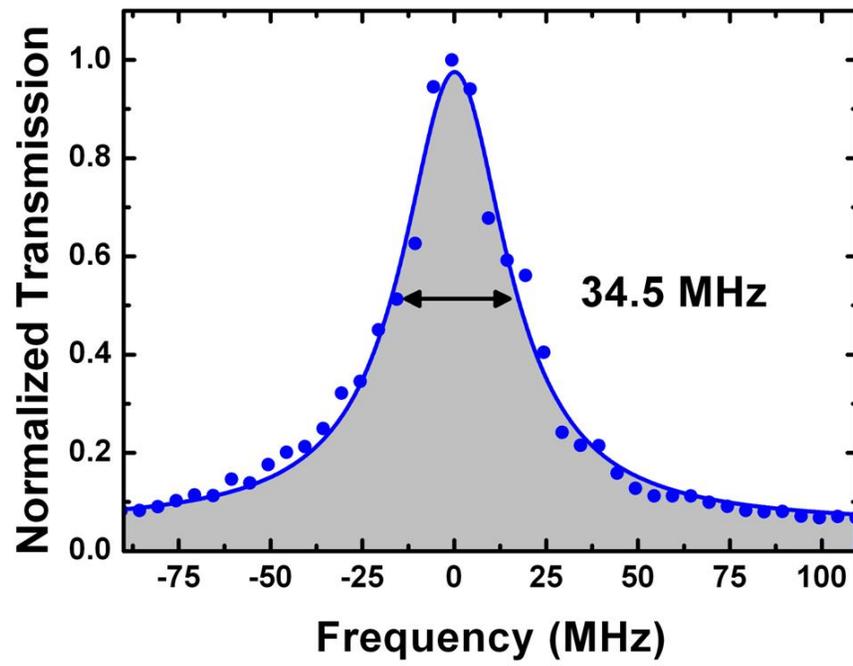


Figure S4:

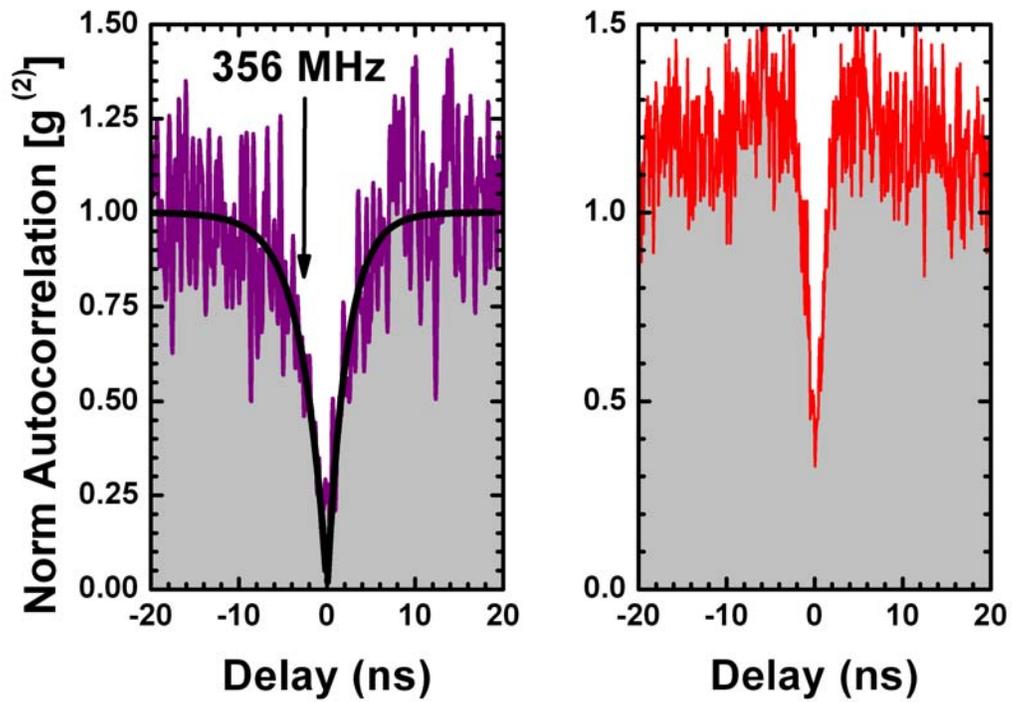


Figure S5:

