Supplementary material for "Current-injection quantum-entangled-pair emitter using droplet epitaxial quantum dots on GaAs(111)A"

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In this supplementary material, we report an atomic microscope image of the quantum dot surface (Suppl. Fig. 1), the detailed layer sequence of our diode samples (Suppl. Fig. 2), discussion about the device characteristics at low temperatures, and a complete set of coincidence histograms for all polarization combinations (Suppl. Fig. 3).

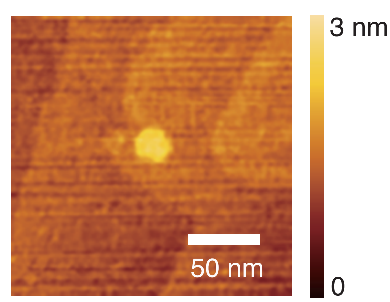
Supplementary Discussion

Diode characteristics at low temperatures. Figure 1(c) in the main body of this paper shows the EL intensities of the X and XX spectral lines as a function of bias current I for temperatures of 10 and 70 K. The impact of temperature on the EL intensities is summarized as follows: 1) The saturation intensity is roughly four times lower at 70 K than 10 K. A similar behavior is also seen in Fig. 1(b). This is due to the thermal escape of charge carriers out of quantum dots. Since our dots have a shallow quantum confinement as low as 50 meV, the EL intensity is affected by carrier escape over the present temperature range. 2) With increasing I from zero, the X line at 70 K starts to appear when $I \sim 0.2$ mA, and it reaches the saturation value when $I \sim 2$ mA. In contrast, the X line at 10 K starts to appear when $I \sim 5$ mA, more than ten times higher than the relevant value at 70 K, and reaches saturation when $I \sim 20$ mA, again, ten time higher than the value at 70 K. Thus, we conclude that, at 10 K, we need more than ten times higher injection to obtain the EL intensity similar to that at 70 K. This finding contradicts a common assumption that the probability of electron-hole recombination is proportional to injection current. The mechanism behind this observation is related to the quenching of the free carrier concentration in the n-doped AlGaAs barrier.

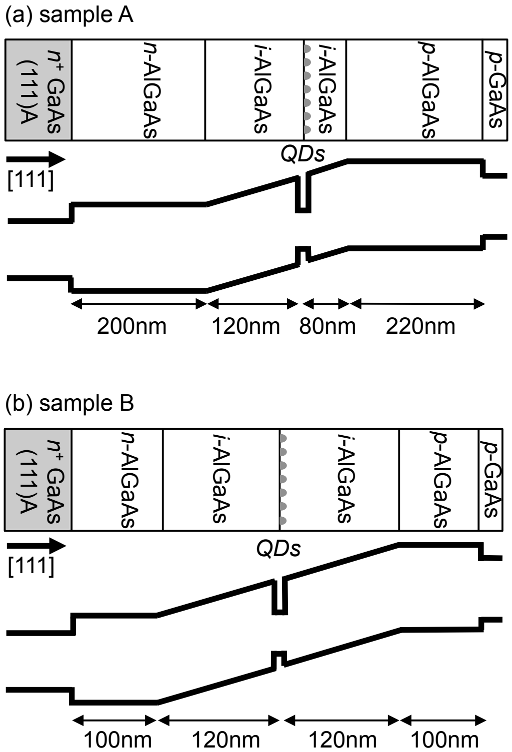
It is known that in n-doped III-V alloy semiconductors, which include Si doped $Al_xGa_{1-x}As$, deep donor levels, often called DX centers, are formed. It is believed that the DX level is a state of the isolated substitutional donor atom [1]. For Si doped $Al_xGa_{1-x}As$ grown by molecular beam epitaxy, the donor activation energy E_D abruptly increases with AlAs mole fraction x, and reaches 150 meV at $x = 0.36$, which is close to the direct-indirect crossover point ($x = 0.42$) [2]. Hence, the free carrier concentration is expected to be seriously quenched at very low temperatures. Note, we choose $x = 0.25$ for the barrier layers in order to enhance the electron concentration, though such a small mole fraction leads to shallow quantum confinement. Nevertheless, the free electron density in the n-doped layer is not so high at temperatures as low as 10 K. In this case we expect that current flow across the intrinsic region in our p-i-n diode is governed by the conductance of holes, which are injected from the p-doped region, passed through the i-region, and recombined with electrons at the interface region between the i- and weakly n-doped regions. Thus, electron-hole recombination can hardly occur in dots unless sufficiently large current is injected. At temperatures as high as 70 K, free electrons are activated, and recombination occurs in dots even with a relatively low bias condition.

This hypothesis is consistent with the following independent observations; (i) The X (XX) intensity dependence on bias current at 10 K deviates from, and is significantly lower than, the expected linear (quadratic) dependence at the low bias regions. (ii) The EL spectra exhibit a strong X^+ line with a negligible X^- line, implying that charge neutrality is broken in the dots, which are mostly occupied by holes.

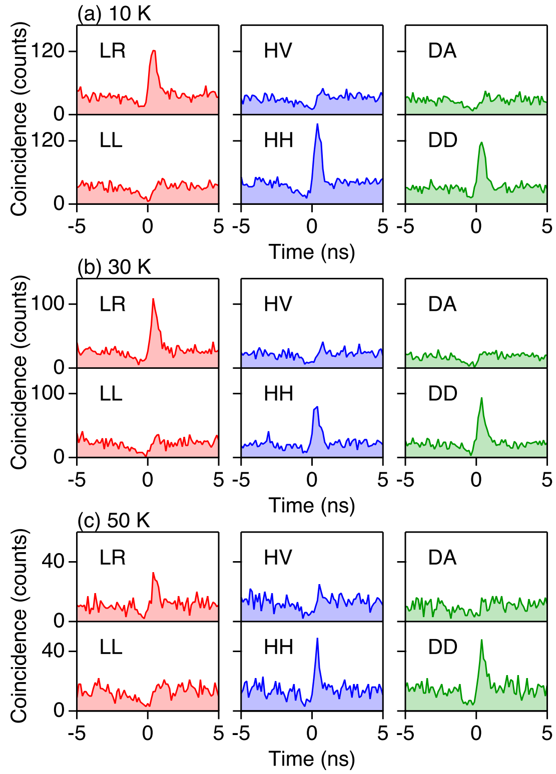
[1] P. M. Mooney, "Deep donor levels (DX centers) in III-V semiconductors," J. Appl. Phys. **67**, R1 (1990).
[2] T. Ishibashi, S. Tarucha, and H. Okamoto, "Si and Sn doping in $Al_xGa_{1-x}As$ grown by MBE," Jpn. J. Appl. Phys. **21**, L476 (1982).



Supplementary Figure 1. Atomic force microscopy image of a quantum dot grown on GaAs(111)A by droplet epitaxy. The dots have a disk-like shape with an average base diameter of 40 nm and a height of 1.0 nm. They are distributed on a (111)A surface with a density of 4×10^8 cm^{-2}. In the actual sample, the dot layer is embedded in a p-i-n diode structure.



Supplementary Figure 2. Layer sequence and energy diagram of (a) sample A and (b) sample B.



Supplementary Figure 3. Coincidence histograms for all polarization settings at 10, 30, and 50 K (sample A). These data are used to determine the fidelity to Bell pairs, shown in Fig. 4(d) in the main body of this paper.