

Rate-limited nonradiative recombination at a single point defect revealed by time-resolved cathodoluminescence

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Nonradiative carrier recombination at point defects (PDs) can severely limit the efficiency of semiconductor optoelectronic devices. However, such nonradiative PDs are difficult to study directly because they are atomic scale, do not emit localized luminescence, and are typically buried deep in a semiconductor heterostructure. As a result, their properties are usually inferred from macroscale optical measurements interpreted using the *ABC* recombination model. Here, we map carrier diffusion and nonradiative recombination at *individual* PDs in a single InGaN/GaN quantum well on the nanometer and nanosecond scale using time-resolved cathodoluminescence. Applying a diffusion-recombination model, we extract the room-temperature intrinsic quantum well carrier lifetime (approximately 4.0 ns) and diffusivity ($0.83 \pm 0.05 \text{ cm}^2 \text{ s}^{-1}$), together with the capture cross section of a *single* isolated PD in the well at $6.8 \pm 0.9 \text{ nm}^2$. We show that nonradiative recombination at this PD takes a minimum time of $16 \pm 2 \text{ ps}$. This limitation predicts that the macroscale defect-related nonradiative recombination rate will saturate above carrier densities routinely reached during device operation, a behavior unaccounted for by the standard *ABC* model.

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I. INTRODUCTION

Over the past decade, solid-state lighting has become a firmly established technology due to the exceptional luminous efficacy of white-light sources based on blue light-emitting diodes (LEDs) [1]. Nonetheless, several promising optoelectronic device classes, including deep-ultraviolet LEDs for sterilization and red micro-LEDs for displays, still face substantial limitations in internal quantum efficiency and reliability which prevent them from reaching wide application [2–4]. In these III-nitride-based devices, and in a range of related optoelectronic systems, crystallographic defects located within or near the quantum well (QW) active region are widely regarded as a major factor limiting performance [5–7]. When acting as efficient Shockley-Read-Hall

(SRH) centers, such defects can enhance nonradiative recombination rates and dramatically reduce the internal quantum efficiency of devices [8].

While past work has often concentrated on extended defects—in large part because their micron-scale signatures are accessible to various microscopy techniques [9–12]—point defects (PDs) are increasingly recognized as key SRH centers in III-nitride devices [13–16]. However, nonradiative PDs buried in QWs are substantially more difficult to study: their atomic scale, often intrinsic nature (e.g., vacancies), and lack of localized luminescence render most structural, elemental, and super-resolution microscopy techniques ineffective. Consequently, PD investigations have principally relied on macroscale optical measurements (e.g., time-resolved photoluminescence) that are frequently analyzed using the *ABC* model [17–20]: $R(n) = An + Bn^2 + Cn^3$, where R is the total carrier recombination rate, n is the carrier density, A is the monomolecular recombination coefficient commonly associated with defect SRH recombination, B is the radiative recombination coefficient, and C is the Auger-Meitner recombination coefficient. Such macroscale analyses infer trends in PD properties and densities from extracted A coefficients, potentially obscuring or averaging out crucial details of the underlying recombination

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physics. By contrast, nanoscopic studies capable of directly capturing such details by resolving and quantifying nonradiative recombination at individual PDs remain scarce. Here, cathodoluminescence (CL) mapping has recently emerged as a promising method since it can provide sufficient spatial resolution to correlate PD locations and QW luminescence under suitable conditions [21,22].

In this work, we use time-resolved CL (TRCL) mapping to follow the nanometer- and nanosecond-scale evolution of carrier diffusion and nonradiative recombination at individual PDs in an InGaN/GaN single QW. By developing a time-dependent diffusion-recombination model, we analyze the nonradiative impact from a *single* PD, explain features in the local QW luminescence decay that fall outside the *ABC* description, and directly derive the PD capture cross section together with the local intrinsic carrier lifetime and diffusivity. Crucially, our analysis shows that recombination at the PD is rate-limited, taking a minimum time of around 16 ps. When extended to uniform-injection conditions, this behavior predicts saturation of the SRH recombination rate above carrier densities on the order of 10^{19} cm^{-3} , a regime routinely reached during typical III-nitride LED operation [23]. Such rate saturation is incompatible with the standard *ABC* model, in which the SRH recombination term An increases with carrier density without limit.

II. SAMPLE AND INITIAL CL CHARACTERIZATION

III-nitrides provide an ideal platform to study PD non-radiative recombination in QWs. Recent studies have identified an intrinsic mid-gap PD type in InGaN/GaN QWs, likely a nitrogen-indium divacancy, that acts as a strong nonradiative recombination center [15,16,21,24,25]. Recombination at these PDs is fully nonradiative within experimental sensitivity, with no detectable defect luminescence even at PD densities above 10^{16} cm^{-3} in the QW (see Appendix A). Furthermore, the density of these PDs can be precisely controlled via an indium-containing “underlayer” grown below the QW [24]. However, many of the details of this type of III-nitride PD, including its capture cross section, remain unclear.

The InGaN/GaN single QW sample studied here is depicted in Fig. 1(a). The sample was engineered for a minimal PD density to give the best chance of spatially resolving recombination at a single PD by using a thick PD-density reducing underlayer (see Appendix B 1), as well as an ultrathin QW to further suppress the number of PDs per QW unit area. This approach yielded a key nonradiative PD density of approximately $1 \mu\text{m}^{-2}$ experienced by carriers in the QW, as determined from prior continuous-excitation CL mapping [21] and matched by deep-level optical spectroscopy on a similar sample [26]. The sample is measured at room temperature, and the

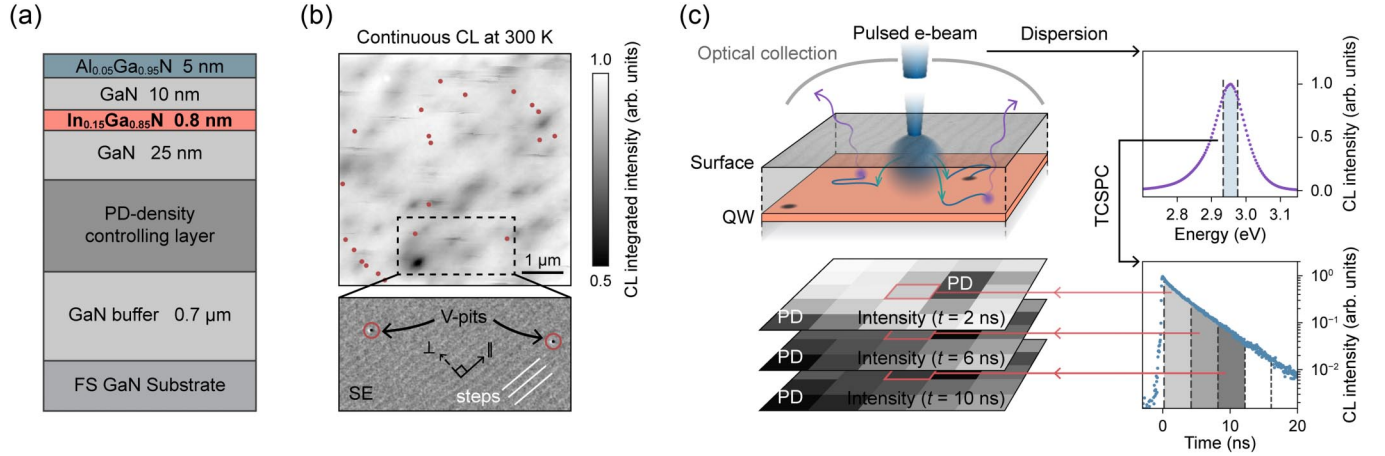


FIG. 1. (a) Schematic of the studied InGaN/GaN QW structure. A thick PD-density controlling layer and thin 0.8 nm $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}$ QW result in a minimal 2D PD density. The thin QW also mostly negates the quantum-confined Stark effect present in many III-nitride QWs, which significantly simplifies the understanding and modeling of the TRCL data (see Appendix C). A 5 nm $\text{Al}_{0.05}\text{Ga}_{0.95}\text{N}$ cap reduces the impact of surface carrier recombination, while using a freestanding (FS) GaN substrate limits the dislocation density to one to two orders of magnitude below the PD density. (b) Integrated QW intensity image acquired from hyperspectral continuous-excitation CL, with a section of the simultaneously acquired secondary-electron (SE) image below. Positions of V-pits on the surface as detected from the secondary-electron image are marked by red dots. No spatial correlation exists between dark spots in CL and the V-pit locations. Directions parallel and perpendicular to the step edges are marked by black arrows. (c) Schematic of TRCL mapping. A pulsed electron beam excites the QW sample. The collected CL signal is dispersed and the emission within a spectral window of the QW peak energy is forwarded to a time-correlated single photon counting (TCSPC) detection system to measure a CL decay curve. The full TRCL decay map is created by repeating the process as the electron beam scans the surface; the 3D dataset can be visualized as a sequence of intensity images by integrating all decay curves over selected time ranges.

excess carrier densities generated in the QW are on the order of those in blue LEDs under operating conditions [23] (10^{19} cm^{-3} , see Appendix D), making our results applicable to real-world devices. In addition, around this carrier density the QW is close to peak radiative efficiency, with previous work estimating the macroscale internal quantum efficiency to be around 60% due to the low PD density [21].

For an initial understanding of the sample luminescence, we inspect an integrated QW intensity image acquired on a $6 \times 6 \mu\text{m}^2$ area from continuous-excitation CL [Fig. 1(b)]. This area of the sample remains the focus of the rest of this work. Each dark spot in the image corresponds to the local enhancement in nonradiative recombination rate from an individual PD, with the density of dark spots at around $1 \mu\text{m}^{-2}$ in agreement with the expected PD density. The dark regions are uncorrelated with the locations of dislocation-related V-pits on the surface, further confirming that they arise from nonradiative PDs rather than extended defects. The dark spots are slightly elongated along the step-edge direction. This shape is due to anisotropic carrier diffusion which originates from step-related QW-energy fluctuations in these thin-QW samples, as evidenced in Refs. [21,27] and further expanded on in Appendix E. This anisotropy is included in the modeling of our TRCL results in Sec. IV.

III. TRCL MAPPING AND POINT-DEFECT ISOLATION

Figure 1(c) shows a schematic of TRCL mapping. An electron pulse locally generates electron-hole pairs, which rapidly relax to the QW, setting up an initial excess carrier density distribution in the well (see Appendix D). The excess carriers then diffuse laterally within the QW, either recombining via radiative/Auger-Meitner processes, or via nonradiative SRH if they diffuse to a PD location. Fully considering carrier diffusion when interpreting CL results is crucial: recent studies have indicated that carrier diffusion coefficients in InGaN/GaN QWs can be as high as $3 \text{ cm}^2 \text{ s}^{-1}$ at room temperature [29,30], contrary to previous conclusions of carrier localization [31–34]. To record the QW-luminescence decay resulting from these recombination pathways, the globally collected CL is dispersed and the emission around the QW peak is directed to a time-resolved detection system (see Appendix B 2). This procedure is repeated for each excitation position to build up the three-dimensional TRCL decay map. We can visualize this 3D dataset by integrating the signal within a series of time windows to generate a sequence of 2D images which show how the QW luminescence intensity evolves over time. As in Fig. 1(b), PDs appear as dark regions in these intensity images because they enhance nonradiative recombination in their vicinity.

Figures 2(a)–2(c) present such TRCL QW intensity images at 0.5, 2.0, and 6.0 ns after excitation. The

evolution of carrier diffusion and nonradiative recombination is directly visible in these images. At 0.5 ns, carriers have had a very short time in which to diffuse, so only a small proportion injected close to the PDs have undergone nonradiative recombination. As such, the low-intensity regions extend only slightly beyond the PD positions, and overall PD contrast in the image is weak. As time progresses, these dark regions grow in size and contrast as carriers diffuse farther from the injection location and recombine at more distant PDs. By $t = 6 \text{ ns}$, carriers have diffused far beyond the average PD spacing, so the effects of adjacent defects overlap.

These images show how the spatial resolution in TRCL degrades in the time, t , after the electron pulse arrives, in-line with the increasing carrier diffusion distance given by $2\sqrt{Dt}$, where D is the carrier diffusivity. In fact, the spatial resolution in TRCL rapidly becomes worse than that of continuous-excitation CL, even when considering times below the intrinsic carrier lifetime (approximately 4 ns for this sample; Appendix D). This counterintuitive effect is clear if we compare Figs. 1(b) and 2(b), and it arises from differences in the carrier-density profiles generated by continuous and pulsed excitation (see Appendix F). We identify $t = 2 \text{ ns}$ as the time beyond which we can no longer discern the impact of *individual* defects on the carrier dynamics in the QW—we exclude TRCL intensity collected after this time from our single-PD analysis.

Next, we isolate the TRCL decays around a single PD [Fig. 2(d)] and further extract data along a single line to simplify modeling [Fig. 2(e)]. To illustrate the variation in decay shape, we plot three representative curves: one for carrier injection directly at the PD, one at an intermediate distance, and one far from the PD which reflects the intrinsic QW decay. The decay curves exhibit faster *initial* decay as the injection position approaches the PD, as evidenced by the increasing deviation from a monoexponential fit to the early decay in Fig. 2(e). This observation is surprising: according to the standard ABC-model interpretation of carrier recombination, SRH recombination should dominate at lower carrier densities and hence have a stronger impact on the luminescence decay at *later* times. To elucidate this phenomenon and glean quantitative information on this single PD, we need to develop a model to track local carrier diffusion and recombination.

IV. SINGLE POINT DEFECT DIFFUSION-RECOMBINATION MODEL

Neglecting drift, the time-dependent excess 2D carrier density in the InGaN/GaN QW, n , follows:

$$\frac{\partial n(x, y, t)}{\partial t} = \left[\nabla \cdot \begin{pmatrix} D_{\parallel} & 0 \\ 0 & D_{\perp} \end{pmatrix} \nabla \right] n(x, y, t) - R(n(x, y, t)), \quad (1)$$

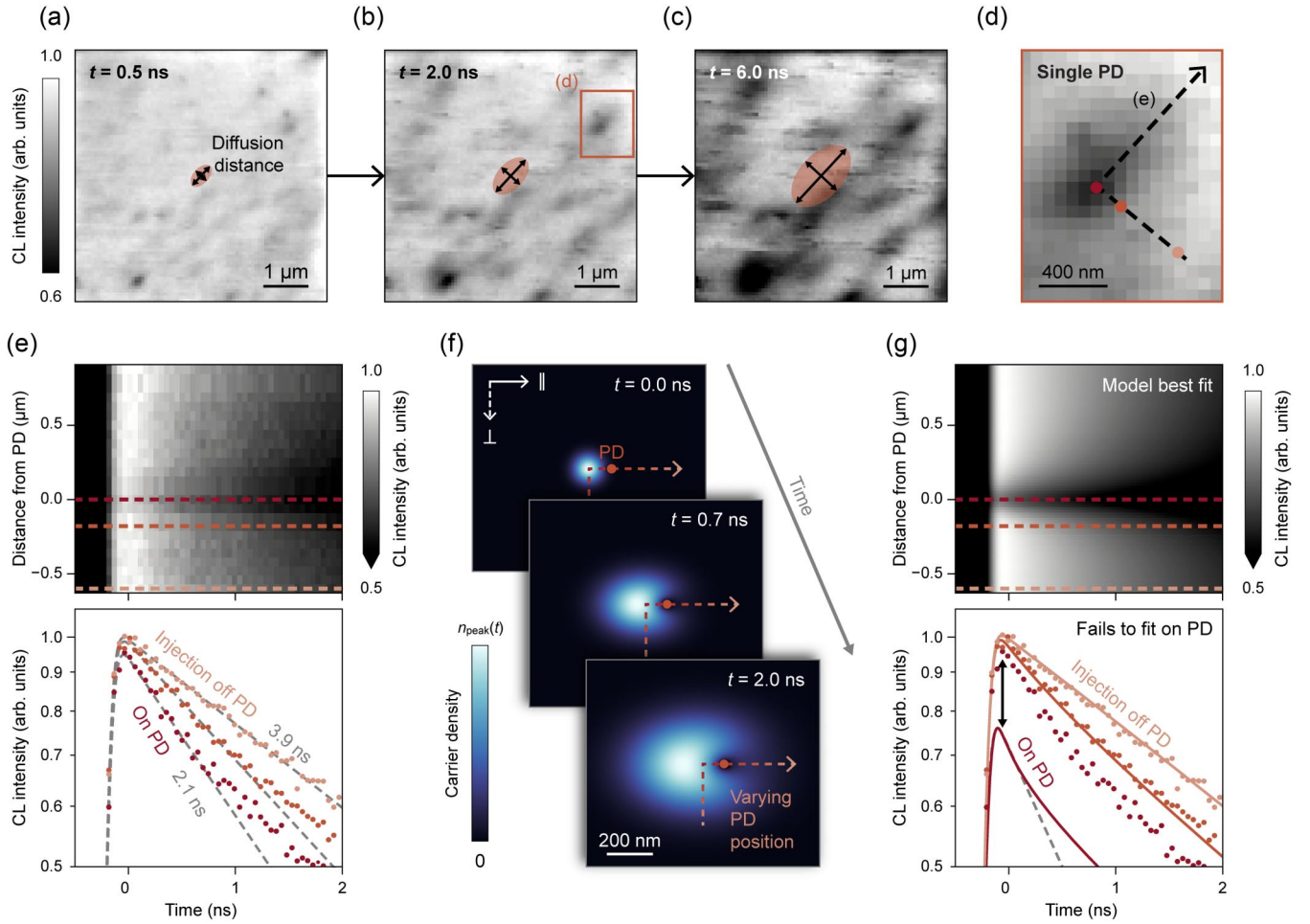


FIG. 2. Time-dependent QW intensity images calculated by integrating the TRCL decay map over time windows of ± 0.5 ns centered on (a) $t = 0.5$, (b) 2, and (c) 6 ns. Each image is individually normalized; see Supplemental Material for an animation of the map at every 0.5 ns interval [28]. Here, $t = 0$ is defined as the time at which the summed TRCL decay curve for the map reaches a maximum (about 0.3 ns after the luminescence initiates). The contrast and area of each dark spot caused by an individual PD increase with time as carriers diffuse farther from the original injection location. Black arrows at the center of each image indicate the diffusion distances calculated from the fitted diffusivities in Fig. 3(c). (d) The response associated with a single PD isolated from the map. The PD position is taken as the location of the pixel with the minimum intensity at $t = 0$. (e) Decay curves extracted along the black line profile, with intensity normalized to the average peak intensity when injecting far from the PD (see Appendix D). The line contains two perpendicular components to capture the full anisotropy in carrier transport. Three curves from this set are plotted below; best monoexponential fits of the early decay are shown in gray (see Appendix B 2 for the fitting equation). Injecting carriers near the PD leads to a faster initial decay as shown by the near 50% decrease in the extracted effective lifetimes (gray values). (f) Example of $n(x, y, t)$ evaluated for a PD-injection position separation of 100 nm along the parallel direction ($\sigma = 1300 \text{ nm}^2$, $D_{\parallel} = 0.08 \text{ cm}^2 \text{ s}^{-1}$, $D_{\parallel}/D_{\perp} = 2.25$, $\tau_{\text{bg}} = 3.9 \text{ ns}$). An animation of the solution is provided in Supplemental Material [28]. Acquiring further solutions while varying the PD location relative to the injection position models the single-PD dataset in (e). (g) Best-fit result of the initial model for the single-PD dataset. The model reproduces the faster initial TRCL decay for carrier injection closer to the PD (monoexponential fit shown as a gray line), but it underestimates the absolute intensity when injecting on the PD location ($\sigma = 6.8 \text{ nm}^2$, $D_{\parallel} = 0.83 \text{ cm}^2 \text{ s}^{-1}$, model results are solid lines).

where t is the time since excitation, x and y are spatial axes aligned parallel and perpendicular to the step edges, $D_{\parallel}$ and $D_{\perp}$ are the ambipolar diffusion coefficients along these directions, and R is the recombination rate. In the absence of the PD, $R(n)$ encompasses all recombination associated with radiative recombination, Auger-Meitner

recombination, or any minor contribution from far less active PDs; we expect radiative recombination to dominate given the high internal quantum efficiency of this QW. These background processes are linked to a position-independent carrier lifetime, τ_{bg} . We treat τ_{bg} and its radiative lifetime, τ_r , as carrier-density independent; this

approximation is justified by the monoexponential decay observed for this QW when exciting far from the PDs (Appendix G).

To model the PD, we assign a cross-sectional area centered on the PD location. This cross section represents the capture probability for whichever type of carrier corresponds to the slower step in the SRH process. That carrier species is then the one tracked by our model, while the other is assumed to instantly complete the SRH recombination cycle. This is a common assumption, since the capture of one type of carrier is usually significantly slower than the other due to the exponential dependence of capture probability on the PD energy level relative to the band edge [13]. It follows that any modeled carrier which strikes this cross section will immediately non-radiatively recombine, imposing a Dirichlet boundary condition with $n = 0$.

The resulting model is described by

$$\frac{\partial n(x, y, t)}{\partial t} = \left[\nabla \cdot \begin{pmatrix} D_{\parallel} & 0 \\ 0 & D_{\perp} \end{pmatrix} \nabla \right] n(x, y, t) - \frac{n(x, y, t)}{\tau_{bg}}, \quad (2)$$

with the boundary condition

$$n(x, y, t) = 0 \quad \text{if } (x - x_{PD})^2 + (y - y_{PD})^2 \leq \frac{\sigma}{\pi}, \quad (3)$$

where x_{PD} and y_{PD} are the center coordinates of the PD while σ is its cross-sectional area. Given constant extraction and collection efficiency, the globally-collected TRCL intensity, $I(t)$, depends on the total number of carriers, n_{tot} :

$$I(t) \propto \frac{n_{tot}(t)}{\tau_r} \propto \iint n(x, y, t) dx dy, \quad (4)$$

where our treatment of τ_r as carrier-density independent let us set the intensity proportional to n_{tot} . Thus, solving Eq. (2) for $n(x, y, t)$ yields the predicted TRCL decay curve.

An example of $n(x, y, t)$ numerically evaluated for a 100 nm separation between the PD and carrier injection location is shown in Fig. 2(f) (see Appendix B 3 for numerical solution details). Spatially summing the solution gives $n_{tot}(t)$, and thus $I(t)$ for this one injection position. To compare the calculation with experiment, $I(t)$ is convoluted with the instrument-response function of our detection system, which has a width of approximately 140 ps. Decays at other injection points are modeled by shifting the PD location while keeping the injection position fixed.

We then fit the model to the single-PD dataset in order to determine the relevant defect and transport parameters. In its most general form, the model depends on five quantities: σ , $D_{\parallel}$, $D_{\perp}$, τ_{bg} , and the initial carrier profile $n(x, y, t = 0)$. However, several of these can be estimated

directly from the measurements or from supporting analysis. In Appendix D, we estimate $\tau_{bg} = 4.0$ ns, find $D_{\parallel}/D_{\perp} \approx 3.0$, and show that $n(x, y, t = 0)$ is well-described by a Gaussian with a full width at half maximum of 70 nm. The fit is therefore reduced to two free parameters, σ and $D_{\parallel}$.

The best-fit result is shown in Fig. 2(g). The modeled curves reproduce the faster initial decay observed experimentally when the injection location is near the PD. This behavior is intuitive in the context of our diffusion model: when carriers are first generated near the PD, nonradiative recombination occurs in a rapid cascade. However, as time passes, the carrier density near the PD decreases as carriers diffuse, and hence the average carrier lifetime increases toward τ_{bg} .

The contradiction between these results and the ABC-model interpretation of decay curves challenges the validity of applying the ABC model to time-resolved measurements. If such experiments are carried out considering (i) a sample with substantial carrier diffusivity and (ii) an initial carrier distribution narrower than the separation between the dominant defects in this sample, we suggest that treating results in the basic ABC framework could lead to severely inaccurate conclusions. We note that these conditions are often reached in TRCL and high-injection time-resolved photoluminescence measurements, which use small laser-spot diameters. Overall, these risks add to mounting critiques of interpreting time-resolved data using the basic ABC model [35,36].

Nevertheless, our initial model is unable to fully reproduce the data [see Fig. 2(g) and Appendix H]. The best fit matches data for injection positions far from the PD, but severely underestimates the peak TRCL intensity when carriers are injected close to the PD. This discrepancy indicates that the model overestimates the initial recombination rate at the PD under those conditions. To grasp the physical mechanism limiting the recombination rate at the PD, we need to consider the SRH process in greater detail.

V. MODEL EXTENSION: POINT-DEFECT DELAY TIME

Figure 3(a) presents an example of the SRH cycle for the recombination of an electron-hole pair at a PD [37]. The cycle consists of four steps: (1) energy-conserving capture of a carrier at the PD, (2) relaxation of the PD in its new charge state via multiple phonon emission, (3) capture of the remaining opposite charge carrier, and (4) further multiphonon emission for the PD to relax to its initial state. Our model above describes one of the capture stages [(1) or (3)] through the diffusion of carriers to the PD cross section. More specifically, only the slower capture stage is included, while the faster one is neglected as discussed in Sec. IV. However, the two multiple phonon emission

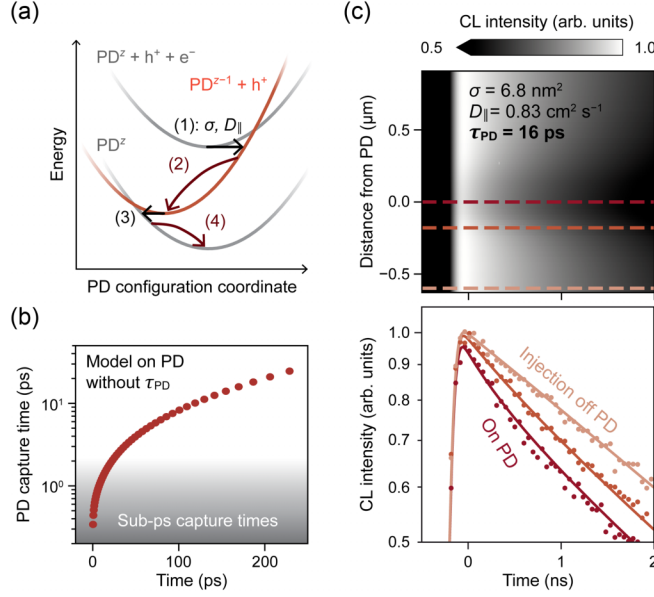


FIG. 3. (a) Schematic of an SRH recombination cycle at a PD with initial charge state z . Our initial model describes one of the carrier capture stages [(1) or (3)] through the D and σ parameters, but it neglects the time required for multiple phonon emission in stages (2) and (4). Our modeling of only one capture stage leads to (b) sub-picosecond PD capture times immediately after carrier injection on the PD. These capture times are close to predicted phonon lifetimes in GaN and therefore leave insufficient time for the multiphonon-emission stages. (c) When we include a PD-recombination delay time, τ_{PD} , in our model to account for stages (2) and (4), the model's best-fit result to the single-PD TRCL dataset improves substantially. The same three decay curves as in Fig. 2(e) are extracted from the experimental data and from the model to display the fit more clearly (model results as solid lines).

events in stages (2) and (4) are not represented in our model—no time is allotted for PD relaxation.

Neglecting the relaxation stage is a common assumption, given that carrier capture should be the much slower part of the recombination cycle. Indeed, this approximation is predicted to be reasonable for excess carrier densities less than 10^{19} cm^{-3} in GaN (around $1 \times 10^{12} \text{ cm}^{-2}$ for our QW) [37]. However, the highly localized excitation of TRCL in our experiment means the initial carrier density state exceeds this limit by roughly a factor of four (Appendix D). Consequently, when injecting the initial state close to the PD location, the assumption of rate-limiting carrier capture breaks down as the PD is subjected to extreme carrier densities.

We elucidate this issue by inspecting the best-fit result from our current model. Focusing solely on the solution for $n(x_{PD}, y_{PD}, t)$ (injecting directly on the PD), we deduce the defect capture times, which represent how long it takes for the PD to capture a single carrier [Fig. 3(b)]. These capture times increase with the time since excitation as carriers diffuse away from the PD. Immediately after excitation,

the predicted capture times are extremely fast at only 300 fs, and they remain well below 10 ps until over 100 ps after excitation. In comparison, longitudinal-optical phonon lifetimes in III-nitride structures start from around 100 fs and can be as long as a few picoseconds [38]. Given that tens of phonons are required to dissipate the QW bandgap energy, it follows that our model is not allowing sufficient time for the PD relaxation to complete at stages (2) and (4); consequently, it fails to match the experimental TRCL decays as in Fig. 2(g).

Accordingly, we replace the original fixed boundary condition around the PD cross section with a set of alternating boundary conditions. The Dirichlet condition in Eq. (3) only holds until the first carrier is captured. At that point, it is replaced by a Neumann condition, which prevents any further carrier flux into the PD cross section:

$$\nabla n \cdot \hat{\mathbf{p}} = 0, \quad (5)$$

where $\hat{\mathbf{p}}$ is the unit normal vector at the circumference of the PD cross section. This Neumann condition persists for a “delay time,” τ_{PD} , which represents the time required for PD relaxation. After that interval, the Dirichlet condition is reinstated until the next carrier capture event occurs; the boundary conditions continue in this loop throughout the numerical evaluation.

Figure 3(c) presents the excellent agreement achieved when this adapted model is fitted to our TRCL dataset with τ_{PD} as the third fitting parameter. The final cross section deduced for the single PD is $6.8 \pm 0.9 \text{ nm}^2$, which places it above most values previously reported for PD cross sections in the III-nitrides [37,39–41]. Only $V_{Ga}V_N$ has been estimated to have a similar cross section [41]. Such a significant cross section confirms the previous identification of this PD type as a critical nonradiative recombination center in InGaN/GaN QWs. Meanwhile, the parallel diffusion coefficient is estimated as $D_{||} = 0.83 \pm 0.05 \text{ cm}^2 \text{ s}^{-1}$, giving the corresponding $D_{\perp} = 0.28 \pm 0.01 \text{ cm}^2 \text{ s}^{-1}$ through the fixed diffusivity ratio. These substantial diffusion coefficients reinforce the case for long-range carrier diffusion in InGaN/GaN QWs and are in fair agreement with previously measured values [29,42]. Finally, our fitted model predicts an intrinsic PD delay time of $16 \pm 2 \text{ ps}$. To our knowledge, this quantity has not been explicitly considered or measured in prior experimental studies of PD recombination and therefore warrants further discussion.

VI. DISCUSSION OF THE POINT-DEFECT DELAY TIME

Our extracted delay time for recombination at a single nonradiative PD provides direct information about a process that is usually very challenging to access experimentally. Yet the fitted value of 16 ps is surprisingly long for a delay governed purely by multiple phonon emission,

given that these events are generally expected to take a few picoseconds at most [37].

A potential source of extra recombination delay is the time required to capture the opposite charge carrier, which is not tracked by our model. However, even in the unlikely case that the capture probabilities of electrons and holes are near equal, accounting for the capture of the opposite carrier would at most double the total PD capture times in our model without the PD delay time. Given that these initially-modeled capture times were as short as a few hundreds of femtoseconds [Fig. 3(b)], such a correction would still leave them far below the 16 ps delay required to explain the observed decay curves near the PD. Consequently, the time needed for the capture of the opposite charge carrier cannot be a significant contribution to the derived PD delay time.

A more likely source of the additional delay is Coulomb repulsion between the PD in one of its charge states and an approaching charge carrier [for instance, if $z = +2$ for hole capture in Fig. 3(a)]. Such repulsion is not included in our model and is accounted for in theory by a scaling factor, referred to as the “Sommerfeld factor,” when calculating capture rates for a charged defect. Overall, our 16 ps PD delay time is likely incorporating the two multiple phonon emission events required for full PD relaxation, as well as any repulsive Sommerfeld effect for the rate-limiting carrier capture event.

Here, we consider the broader implications of this intrinsic PD recombination delay time in a QW. The nonradiative recombination rate associated with a particular defect under uniform carrier injection is usually assumed to be proportional to the carrier density as $N_{\text{PD}}\nu\sigma n$, where N_{PD} is the defect density, and ν is the carrier thermal velocity. This relation underpins the SRH recombination term An in the *ABC* model. However, including a PD delay time imposes a maximum SRH recombination rate of $N_{\text{PD}}/\tau_{\text{PD}}$. This limit defines a critical carrier density n_c beyond which the SRH rate associated with this defect no longer increases:

$$n_c = (\tau_{\text{PD}}\sigma\nu)^{-1}. \quad (6)$$

Notably, while the maximum SRH limit increases with defect density N_{PD} , the critical carrier density n_c is independent of it.

Considering the values of σ and τ_{PD} derived for the single PD in this study, and estimating ν using $\nu = \sqrt{2k_{\text{B}}T/m^*}$ where k_{B} is Boltzmann’s constant, T is temperature, and m^* is the effective mass of the rate-limiting carrier, we calculate $n_c \approx 4 \times 10^{18} - 1 \times 10^{19} \text{ cm}^{-3}$ (around 10^{12} cm^{-2}) for this type of defect depending on whether heavy holes (transverse $m^* \sim 1.8m_0$) or electrons ($m^* \sim 0.2m_0$) are the rate-limiting charge species. This value suggests that the SRH recombination rate could saturate at carrier densities already encountered in

standard LED operation [23], evidencing the importance of the PD delay time to understanding real-world device behavior. Furthermore, it follows that even ignoring the radiative and Auger-Meitner lifetime dependence on carrier density, the relative contribution of PD-related recombination would still decrease under increasing excitation of the QW. This trend represents another departure from behavior predicted by the *ABC* model, adding to the limitations already discussed in Sec. IV.

VII. CONCLUSION

In this study, we acquired a TRCL decay map at 300 K that directly resolves the nanometer- and nanosecond-scale evolution of carrier diffusion and SRH recombination in a high-quality single InGaN/GaN QW. We then isolated and addressed the impact from a single PD in this TRCL map. Using a diffusion-recombination model, we explained the local luminescence decay shape and derived the PD’s capture cross section ($6.8 \pm 0.9 \text{ nm}^2$), as well as the unrestricted carrier diffusion coefficient in the well ($0.83 \pm 0.05 \text{ cm}^2 \text{ s}^{-1}$). Furthermore, we demonstrated that including a recombination delay time for the PD is necessary to explain the TRCL data and proposed that this delay encapsulates the time needed for PD relaxation and any contribution from the Sommerfeld factor. At $16 \pm 2 \text{ ps}$, our derived delay time is long enough that the SRH recombination rate for this defect is predicted to saturate at carrier densities commonly reached during LED operation, a behavior incompatible with the widely used *ABC* model. Consequently, our results show that applying the *ABC* model to macroscale measurements to infer point defect densities and properties should be approached with caution, particularly in the high-injection regime. More broadly, the approach demonstrated here provides a route to compiling statistics of PD capture cross sections and delay times in InGaN QWs and other semiconductor heterostructures.

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J.-F.C. grew the sample. T.F.K.W. and G.K. performed the TRCL measurements. T.F.K.W. was responsible for analysis and modeling carried out on the raw data from the measurements. T.F.K.W., G.K., R.A.O., R.B., and N.G. wrote the paper. T.F.K.W., N.G., and R.A.O.

conceived and directed the project. All authors discussed the results and commented on the manuscript.

DATA AVAILABILITY

The data and code supporting the findings of this study are publicly available [43].

APPENDIX A: NO DETECTION OF DEFECT LUMINESCENCE

We can question whether the critical PDs in InGaN/GaN QWs studied in this work result in any luminescence of their own. Such localized luminescence could be exploited for super-resolution microscopy, as well as providing intrinsic information on the nature of the PDs themselves. However, no PD deep-level luminescence was detected in CL from a sample with a critical PD density $> 10^{16} \text{ cm}^{-3}$ in the QW across the entire wavelength range 600–1700 nm (energy range 0.73–2.1 eV, Fig. 4), even when using integration times as long as 60 s. For more details on this highly defective sample, see Ref. [21]. The absence of detectable luminescence is striking, given that the estimated internal quantum efficiency (IQE) for this sample is only around 1% at room temperature and, as such, the vast majority of injected carriers should be recombining at the PDs. Furthermore, this observation was consistent whether measuring at 10 or 300 K.

Such strongly nonradiative behavior is not entirely surprising: radiative electron/hole capture coefficients for most midgap PDs are orders of magnitude lower than

nonradiative capture coefficients [13]. Nevertheless, we do not rule out any chance of finding luminescence associated with these PDs in the future: perhaps measurements with more precise excitation schemes could find some luminescence signature.

APPENDIX B: METHODS

1. Sample growth

The sample was grown by metal-organic vapor phase epitaxy in a horizontal Aixtron 200/4 RF-S reactor on an n-type *c*-plane freestanding GaN substrate (threading dislocation density approximately 10^6 cm^{-2}). After a low-temperature GaN buffer, 700 nm of GaN is deposited at 1000 °C using trimethylgallium with H_2 as the carrier gas. The temperature is then decreased to 770 °C, and the carrier gas is switched to N_2 for the growth of a lattice-matched $\text{In}_{0.17}\text{Al}_{0.83}\text{N}/\text{GaN}$ superlattice, using trimethylindium and trimethylaluminum for the InAlN and triethylgallium for the GaN. This InAlN/GaN superlattice acts as the PD-density controlling underlayer in the sample [44]. A 10 nm GaN spacer is grown at the same temperature with triethylgallium. Up to this point, the structure is mostly grown with Si doping at around 10^{18} cm^{-3} ; however, the last two periods of the superlattice and the first 5 nm of the following GaN barrier are highly doped with Si ($> 10^{20} \text{ cm}^{-3}$) to screen the electric field arising from the spontaneous polarization mismatch at InAlN/GaN interfaces [21]. The rest of the structure [see Fig. 1(a)] is grown under similar conditions but with no intentional doping. The InGaN/GaN single QW has a nominal indium content of 15%.

2. Cathodoluminescence

We used an Attolight Allalin 4027 Chronos system for continuous CL and TRCL microscopy. The electron gun is triggered using the third harmonic of a Nd:YAG laser (wavelength = 355 nm) with a frequency of 20.15 MHz to achieve well-spaced electron pulses with widths below 10 ps. The compact column of the Allalin system ensures nearly no temporal broadening of the pulse before it strikes the sample. We set the beam energy to 3 keV and installed a 50 μm aperture to suppress the energy dispersion of the electrons. The narrow aperture led to an extremely low probe current of $< 0.8 \text{ pA}$, and hence an average number of electrons per pulse of at most 0.25. It follows that the majority of existent pulses contained only one electron; less than 1 in 5 pulses had two or more electrons in accordance with a Poisson distribution. The dwell time at each pixel in the TRCL map was 2 s. The collected CL was dispersed, and the emission within $\pm 3 \text{ nm}$ ($\pm 21 \text{ meV}$) of the QW peak was directed to a time-correlated single photon counting (TCSPC) system (time resolution approximately 140 ps); this spectral window fully captures the QW CL decay behavior (see Appendix C).

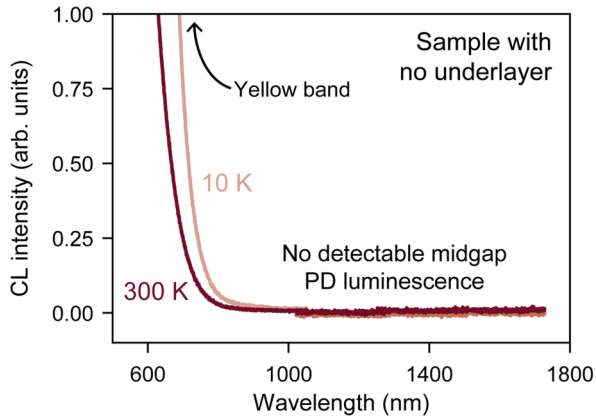


FIG. 4. Continuous CL spectra from a sample with high PD density over a very broad red to infrared region, where Schott glass absorption filters (OG550 and RG850) were applied to block higher order peaks from the GaN and QW emission. Both back-illuminated charge-coupled device and InGaAs photodiode array sensors were employed (above 80% quantum efficiency in the ranges 400–900 nm and 950–1600 nm, respectively), along with diffraction gratings which possessed blaze wavelengths at 500 and 1200 nm. Integration times were fixed to 60 s. No midgap PD emission is present across the entire spectral range, regardless of temperature.

The TRCL decay curves in Figs. 2(e) and 2(g) were fitted using the empirical equation

$$I(t) = \frac{I_0}{2} \exp\left(\frac{-(t-t_0)}{\tau_{\text{eff}}}\right) \left[1 + \operatorname{erf}\left(\frac{2\sqrt{\ln(2)}(t-t_0)}{\Gamma_{\text{IRF}}}\right)\right] + C, \quad (\text{B1})$$

to give the gray dashed lines. This empirical equation is simply an exponential decay convoluted with a Gaussian, which approximates the detection response function. Only data above a relative intensity of 0.8 were fitted to ensure the equation parameterized the early decay behavior in particular. In this equation, τ_{eff} is the “effective” or instantaneous luminescence lifetime, I_0 is the initial intensity, t_0 is a time offset, C accounts for the background noise, and Γ_{IRF} is the detection response function at 140 ps. The resulting values of τ_{eff} are printed in the figure.

3. Numerical evaluation details

Equation (2) was solved for $n(x, y, t)$ under the boundary conditions in Eq. (3) and Eq. (5) using an implicit finite-volume numerical method implemented with the Python package *FiPy* [45].

For the fitting process, we integrated the numerical evaluations into a standard Levenberg-Marquardt fitting loop by automatically generating the finite-volume mesh based on the two fitting parameters, σ and $D_{\parallel}$. The injection location was always fixed to the center of the mesh, as this scheme minimizes the mesh size required to prevent significant carrier densities from diffusing to the border and hence led to reduced computation times. The mesh area was defined as an ellipse with major axis a and minor axis b set to ensure carriers do not diffuse to the simulation boundary within the 2 ns upper time limit, t_{max} :

$$a = 4\sqrt{D_{\parallel}t_{\text{max}} + \Gamma_{\text{init}}}, \quad b = 4\sqrt{D_{\perp}t_{\text{max}} + \Gamma_{\text{init}}}, \quad (\text{B2})$$

where Γ_{init} is the full width at half maximum of the initial carrier density distribution $n(x, y, t = 0)$ (see Appendix D). Neumann zero-flux boundary conditions were always imposed on the boundaries of the mesh area. Mesh cell sizes were set to $\sqrt{\sigma/(4\pi)}$ near the PD cross section and $\Gamma_{\text{init}}/6$ near the carrier injection location to ensure the mesh could fully capture PD boundary conditions and the shape of the initial carrier density distribution, before smoothly increasing to $b/10$ at the edges of the mesh area. An example of the resulting mesh is provided in Appendix D.

APPENDIX C: LIMITED QCSE AND TRCL COLLECTION SCHEME

In Appendix B, we commented that the ± 3 nm spectral window used in our TRCL-TCSPC detection scheme fully

captures the QW luminescence decay behavior for the studied sample. Here, we detail the logic behind this statement.

There is some cause to be wary of a limited spectral window around the QW emission peak for TRCL detection. The partial differential equation used in our recombination-diffusion model only holds if the probability of collecting a photon from a radiative recombination in the QW is independent of the recombination position and the photon energy. The former condition is satisfied by the global collection optics in CL microscopy, but the latter is more problematic due to this spectral detection window. It follows that any strong dependence of the photon energy on the recombination position or time could invalidate our model. This risk is particularly relevant for InGaN/GaN QWs since the quantum-confined Stark effect (QCSE) is known to cause a spectral shift over time in time-resolved measurements due to carrier-density-dependent field screening [46].

Fortunately, QCSE is negligible in our ultrathin QW and hence, there is no observable spectral shift over time as apparent from time-resolved photoluminescence (TRPL) results [Fig. 5(a)]. We confirm this holds in TRCL by sweeping the ± 3 nm TCSPC detection window across the full QW emission wavelength range. The curve acquired by summing all the decay curves across the full QW emission shows exactly the same behavior as the curve acquired from only a single window around the peak QW intensity [see Fig. 5(b)].

We also rule out any effect from position-dependent photon energies: Fig. 5(c) quantifies the change in TCSPC-collected signal due to QW spectral shift across the single-PD path in Fig. 2(d) of the main text. The collected signal varies by less than $\pm 0.5\%$ across the full path, which is insignificant compared to the approximately 25% drop in initial TRCL intensity when injecting carriers at the PD location. The variation is this minor because the peak emission wavelength of the QW at 300 K varies by at most 1 nm across the path, which is negligible compared to the 6 nm TCSPC detection window. Consequently, we are confident that the decay in the measured TRCL signal truly monitors the decrease in total QW carrier population, rather than any carrier transfer to different energy states—this validates our applied model even when detecting within a spectral window.

Furthermore, the almost complete lack of QCSE in this QW also significantly simplifies modeling. Strong QCSE can also cause carrier drift in nonuniform carrier density experiments since high carrier densities locally screen the electric field and generate a secondary electric field in the plane of the well. The negligible QCSE in our sample means we do not need to consider these drift effects in the model used in our main text. (We note these drift effects were still found to be nearly negligible even in thicker InGaN/GaN QWs with stronger QCSE [42]).

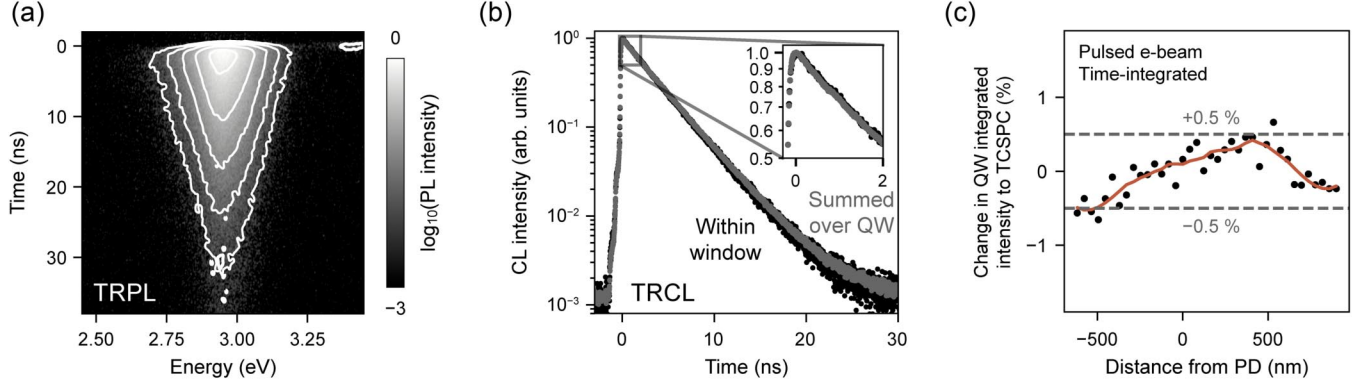


FIG. 5. (a) Streak image acquired using TRPL (excitation wavelength = 266 nm), showing the absence of any QCSE-related spectral shift over time. The TRPL pulse energy density is around $5 \mu\text{J cm}^{-2}$, leading to an initial carrier density in the QW of around 10^{12} cm^{-2} —this value matches our TRCL measurements in the main text. (b) TRCL decay curve calculated by summing many curves gathered across the full QW emission range compared to the decay curve from only detecting around the peak wavelength. Both are identical. (c) Relative change in signal lying within the TCSPC spectral detection window across the single PD path in Fig. 2(d) of the main text. The data are based on time-averaged hyperspectral data gathered by forwarding the dispersed TRCL signal to a charge-coupled device rather than the TCSPC system. At each position, the signal integrated within the globally fixed $\pm 3 \text{ nm}$ TCSPC detection window is compared to the signal within a $\pm 3 \text{ nm}$ window centered on the local peak emission wavelength. Any shift in the local peak emission wavelength away from the global average causes a delta between these two quantities; this delta is plotted on the y axis.

APPENDIX D: EVALUATING FIXED PARAMETERS FOR THE MODEL

There are six parameters involved in the final diffusion model in the main text: the PD cross section (σ), the carrier diffusivities ($D_{\parallel}$ and $D_{\perp}$), the PD delay time (τ_{PD}), the background lifetime (τ_{bg}), and the initial carrier density state [$n(x, y, t = 0)$]. Figure 6 details how we estimated τ_{bg} , $D_{\parallel}/D_{\perp}$, and $n(x, y, t = 0)$ to leave us with only three remaining fitting parameters in our final model to avoid overfitting the data (σ , $D_{\parallel}$, and τ_{PD}).

By averaging TRCL decay curves far from the PD location [blue line in Fig. 6(a)], we can derive the average local decay curve in the absence of any impact from the single PD [Fig. 6(b)]. Fitting this data with Eq. (B1) extracts a value for τ_{bg} at 4.0 ns. The excellent fit of this curve by the monoexponential decay equation across many orders of magnitude in CL intensity demonstrates that τ_{bg} is essentially independent of the carrier density (see Appendix G). The maximum intensity of this average decay curve was also used to normalize all of the decay curves in the main text.

To determine the ratio between the diffusion coefficients, we inspect the shape anisotropy of the dark spot in the $t = 2 \text{ ns}$ image more closely (see Appendix E for an explanation of the shape anisotropy). Fitting the CL intensity along the selected PD path parallel and perpendicular to the step edges [red line in Fig. 6(a)] with an asymmetric Gaussian function extracts two standard deviations, $\sigma_{\text{d},\parallel}$ and $\sigma_{\text{d},\perp}$ [Fig. 6(c)]. We expect that these standard deviations are proportional to the underlying

carrier diffusion distances along the parallel and perpendicular directions given by $2\sqrt{Dt}$, and hence, we derive

$$\frac{D_{\parallel}}{D_{\perp}} = \left(\frac{\sigma_{\text{d},\parallel}}{\sigma_{\text{d},\perp}} \right)^2. \quad (\text{D1})$$

Consequently, we fix $D_{\parallel}/D_{\perp} = 3.0$ from the results of this fit.

To predict the initial state, $n(x, y, t = 0)$, we first calculate the energy density deposited by the electron beam in our sample, ρ_{E} . As mentioned in Appendix B, a single pulse from the beam under our conditions on average contains only a single 3 keV electron. The energy deposited from this electron is calculated using CASINO [47], from which we can estimate the generated carrier density using the commonly applied expression [48]

$$n(x, y, z) = \frac{\rho_{\text{E}}(x, y, z)}{3E_{\text{g}}(z)}, \quad (\text{D2})$$

where z is the depth in the sample and E_{g} is the material bandgap in the heterostructure at that depth. The probe diameter on the sample surface used in the CASINO simulation is estimated at 20 nm by inspecting the secondary-electron image resolution under our measurement conditions, while $E_{\text{g}}(z)$ for our heterostructure is calculated using the bandstructure parameters for III-nitride materials in Ref. [49].

The resulting generated carrier distribution is displayed in Fig. 6(d) (left). However, to predict the true initial state in the InGaN QW we still need to account for extra

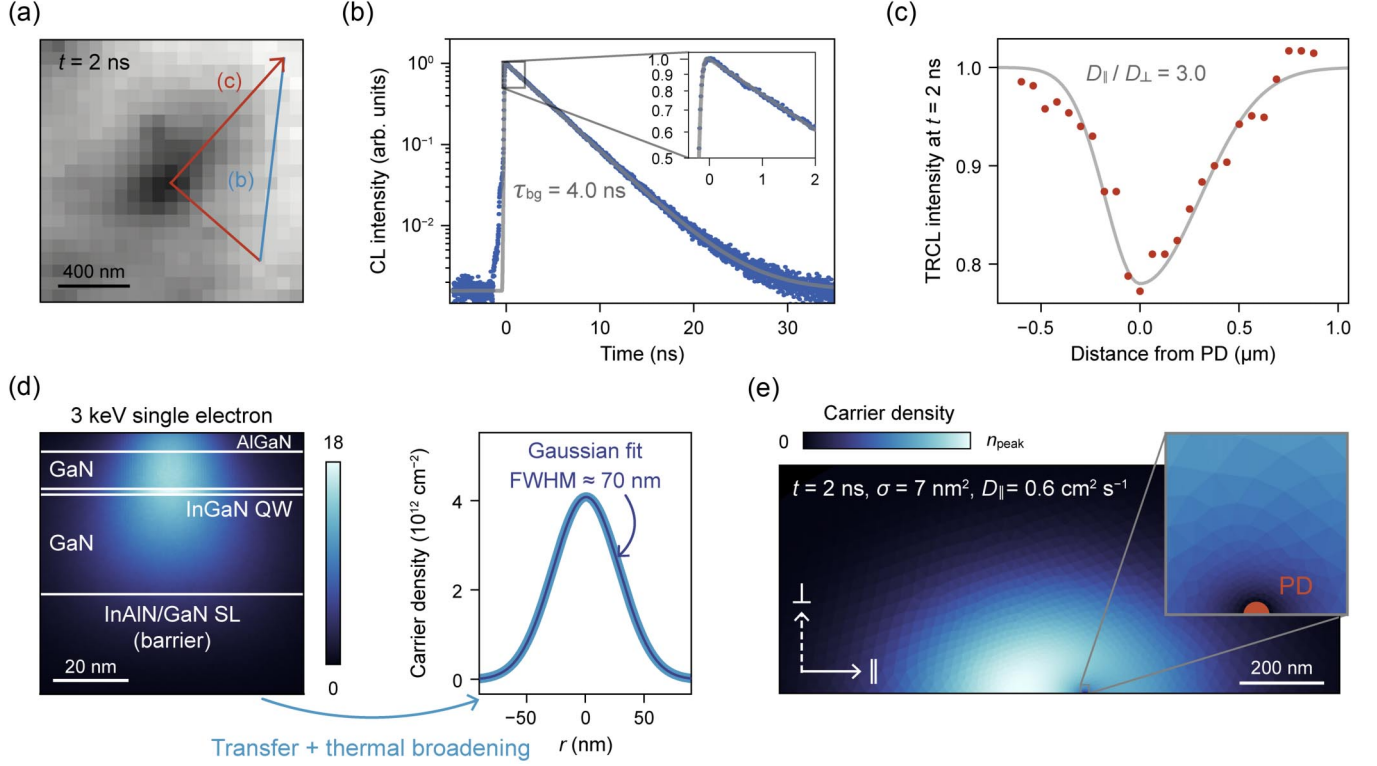


FIG. 6. (a) A section of the TRCL map at $t = 2$ ns as in Fig. 2(d) of the main text, indicating the paths used in sub-figures (b) and (c). (b) Average TRCL decay curve along the blue line in (a), far from the PD location. A fit with Eq. (B1) is shown in gray, extracting $\tau_{bg} = 4.0$ ns. (c) TRCL intensity at $t = 2$ ns along the red path in (a). Fitting the data with an asymmetric Gaussian determines $D_{\parallel} / D_{\perp}$. (d) Left: The carrier distribution in the sample from one incoming 3 keV electron estimated using *CASINO* (integrated over the out-of-page direction, resulting in final units of cm^{-2}). Accounting for thermal and transfer broadening leads to the calculated initial carrier density state in the QW for our model (right), plotted as a function of the radial distance, r , from the excitation position. This state is well-described by a Gaussian distribution with a full width at half maximum (FWHM) of 70 nm. The small proportion of carriers generated in the PD-controlling InAlN/GaN superlattice (SL) layer is excluded from the initial state since the bandgap of $\text{In}_{0.17}\text{Al}_{0.83}\text{N}$ is larger than that of GaN, preventing carriers from transferring to the QW. (e) An example of the automatically generated finite-volume mesh used to evaluate and fit our model to TRCL data.

broadening from (i) the lateral diffusion of the carriers as they transfer from the surrounding higher-bandgap layers to the well, and (ii) the continued diffusion of carriers while they thermalize to lower-energy states, during which they are very unlikely to recombine [50]. We can account for both of these effects by convoluting the *CASINO*-predicted carrier density with a Gaussian, as described in Ref. [21]. The final predicted initial carrier density distribution in the QW is shown on the right of Fig. 6(d) and is well-described by a Gaussian with a full width at half maximum of 70 nm. The carrier density peaks at $4 \times 10^{12} \text{ cm}^{-2}$ (around $4 \times 10^{19} \text{ cm}^{-3}$). As we mentioned in the main text, this value is comparable to densities reached under LED operating conditions and also exceeds the limit above which the relaxation stage in the SRH cycle might reasonably be neglected [37].

Modeling this initial state as instantaneous at $t = 0$ is reasonable given the speed of the processes involved: (i) the electron pulse width is only 10 ps, (ii) the hot-carrier

thermalization time in GaN is expected to be much less than 1 ps [51], and (iii) the carrier transfer from the surrounding GaN barriers to the QW is predicted to take at most 4 ps considering the bulk GaN diffusion coefficient of approximately $0.8 \text{ cm}^2 \text{ s}^{-1}$ at 300 K [52]. All of these time scales are far faster than the 140 ps response of the TCSPC system, and hence, the TCSPC response fully determines the detected luminescence rise time. Furthermore, though the time scales for these initialization processes are on the order of τ_{PD} , they only occur once at the start of the carrier response, while the delayed PD capture process impacts the modeled carrier dynamics up to hundreds of picoseconds after excitation [see Fig. 3(b) of the main text]. Consequently, we expect initialization time to have a negligible influence on our estimation of τ_{PD} .

Finally, it is worth acknowledging that our extracted value for τ_{PD} is sensitive to the *number* of carriers in this initial state, $n_{\text{tot}}(t = 0)$. This sensitivity may induce uncertainty in τ_{PD} which is challenging to quantify, since

our value of $n_{\text{tot}}(t=0)$ is only a prediction. Nonetheless, we stress that the need to introduce τ_{PD} to accurately model the TRCL data is unaffected by this concern, since the model without τ_{PD} does *not* depend on $n_{\text{tot}}(t=0)$, but rather only on the full width at half maximum of $n(x, y, t=0)$. Consequently, even if our value for the delay time possesses some uncertainty, our results clearly demonstrate that the recombination rate at the PD is limited by PD relaxation and/or a repulsive Sommerfeld effect at high enough carrier densities.

Figure 6(e) presents an example of the finite-volume mesh automatically generated during the fitting process to evaluate $n(x, y, t)$ (see Appendix B). Due to the mirror symmetry of the problem for any given PD location along the path, we only need to simulate half of the QW plane, which saves computation time.

APPENDIX E: STEP EDGES AS A SOURCE OF ANISOTROPIC CARRIER DIFFUSION

In the main text, we noted that dark regions due to PDs in continuous CL and TRCL were elongated along the step-edge direction due to anisotropic carrier diffusion. This anisotropic behavior was accounted for in our model by using two diffusion coefficients, $D_{\parallel}$ and $D_{\perp}$.

Figure 7 demonstrates the source of the anisotropic carrier diffusion using CL and scanning transmission electron microscopy (STEM) results. Bilayer-height atomic step-edges on the surface spatially correlate with periodic 5 meV fluctuations in the QW peak CL energy at 10 K [Figs. 7(a) and 7(b)]. STEM cross-sectional imaging in Fig. 7(c) reveals that these energy fluctuations are due to

atomic-bilayer thickness fluctuations in the QW. These thickness variations arise from the step-flow growth mechanism of the QW and were experimentally determined to increase the carrier confinement energy by up to 60 meV at the narrowest section of the well (see Ref. [27] for further detail). Consequently, this sample has an asymmetric in-plane energy landscape: carriers face an extra barrier to diffusion in the direction perpendicular to the step edges, but no barrier parallel to the step edges. This landscape causes anisotropic carrier diffusion, which is maintained up to room temperature, as expected from the fact that the 60 meV activation barrier perpendicular to the step edges is greater than $k_{\text{B}}T$ at 300 K (26 meV). Full temperature-dependent diffusion studies are in Refs. [21,27]. We note that the anisotropic diffusion in this sample is exacerbated by its ultrathin QW; InGaN/GaN samples with thicker QWs are unlikely to show such strongly anisotropic carrier dynamics.

APPENDIX F: CL SPATIAL RESOLUTION

On inspecting the time-dependent CL intensity images generated from the decay map in the main text, we noted that their spatial resolution rapidly became worse than that for continuous-excitation CL, even when considering times below the intrinsic carrier lifetime. We can quantify this effect by comparing the steady-state QW carrier distribution expected from continuous excitation with the time-dependent solution for pulsed excitation in TRCL.

We calculate the time-dependent carrier distribution in TRCL by solving Eq. (2) for $n(x, y, t)$ in the absence of any PD. Meanwhile, the steady-state carrier distribution in

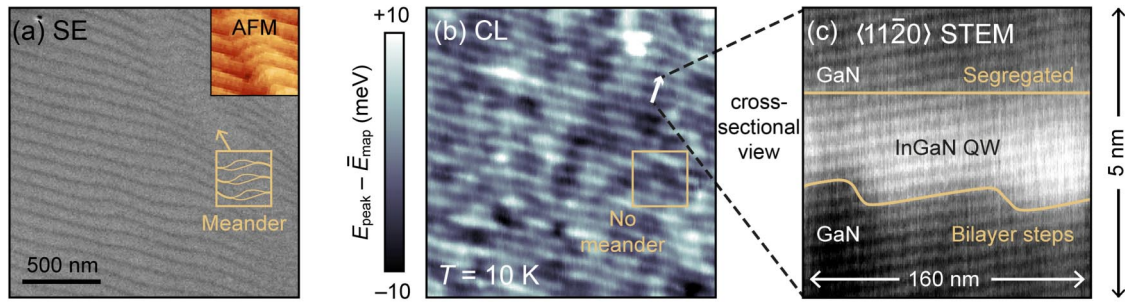


FIG. 7. (a) A secondary-electron (SE) image of the surface of the sample studied in the main text. Atomic bilayer-height step-edges appear as dark-contrast parallel lines, while a step-meandering area is highlighted in yellow. The inset shows an example of such a step-meander in atomic-force microscopy (AFM), for which bilayer steps delaminate into two monolayer steps along one ribbon (AFM is at the same lateral scale as the SE image, height scale 1 nm) [53]. A continuous CL hyperspectral map of this same area is used to generate (b), the corresponding image of the peak QW emission energy at 10 K relative to the average peak energy for the entire area, $E_{\text{peak}} - \bar{E}_{\text{map}}$. Step edges clearly induce a periodic energy modulation across the QW; the absence of any meandering feature rules out surface effects. (c) An atomic-resolution STEM high-angle annular dark field cross section looking down the step-edges (a axis $\langle 11\bar{2}0 \rangle$). Bilayer fluctuations from step-flow growth are visible in the abrupt lower QW interface, while the top interface is segregated. The STEM image has been compressed in the horizontal direction to allow for the extreme differences in scale between step separation (approximately 100 nm) and monolayer height. The lateral scale of the STEM image is indicated for comparison purposes on (b) by a white arrow; the STEM image was not acquired on this exact area.

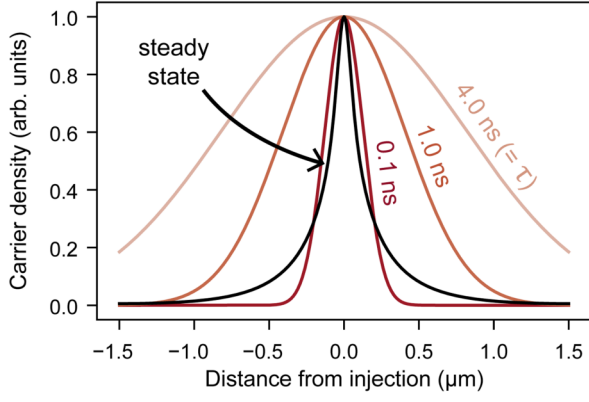


FIG. 8. The steady-state carrier distribution in continuous-excitation CL is compared to time-dependent solutions for the pulsed excitation in TRCL. Both are normalized to the maximum carrier density, and the x axis is aligned along the step-edge direction. The pulsed solution is broader than the steady state, even considering times far below the carrier lifetime.

continuous CL is predicted by solving a similar equation with a generation rate term, $G(x, y)$, and setting $\partial n / \partial t = 0$:

$$G(x, y) = \frac{n(x, y)}{\tau_{bg}} - \left[\nabla \cdot \begin{pmatrix} D_{\parallel} & 0 \\ 0 & D_{\perp} \end{pmatrix} \nabla \right] n(x, y). \quad (\text{F1})$$

The parameters used for these evaluations match the properties of our sample derived in the main text and Appendix D ($\tau_{bg} = 4.0$ ns, $D_{\parallel} = 0.83$ cm² s⁻¹, $D_{\parallel}/D_{\perp} = 3.0$). Similarly, since we used the same electron beam energy in both TRCL and continuous CL, the generation distribution is set to a Gaussian with full width at half maximum of 70 nm for both; in the continuous-excitation case, this Gaussian defines $G(x, y)$, while in the pulsed-excitation case it defines $n(x, y, t = 0)$.

The resulting carrier density solutions are displayed in Fig. 8 and elucidate the source of poor spatial resolution in TRCL. The carrier distribution from pulsed excitation surpasses the width of the corresponding steady-state distribution incredibly quickly, even when considering times far below the total lifetime of the carriers. This sudden spatial resolution loss represents a drawback to TRCL applied to systems with substantial carrier diffusion. For TRCL spatial resolution to surpass continuous CL resolution in these systems, we must consider times over an order of magnitude shorter than the carrier lifetime.

APPENDIX G: CARRIER-DENSITY INDEPENDENT LIFETIMES

In the main text, we treated the carrier radiative lifetime, τ_r , and the background lifetime away from the PD, τ_{bg} , in our ultrathin QWs as carrier-density independent. This treatment allowed us to set the predicted TRCL intensity as

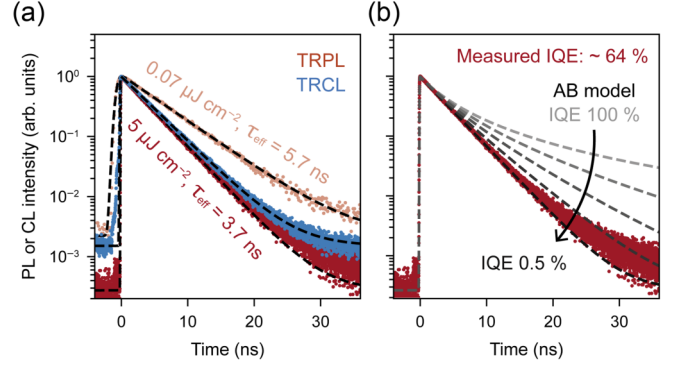


FIG. 9. (a) Room-temperature decay curves of QW luminescence in TRPL and TRCL for the studied sample. The entire curves are well-fitted by a simple monoexponential decay [Eq. (B1)]. TRPL curves for two different excitation densities are included (corresponding to initial carrier densities on the order of 10^{11} cm⁻² and high 10^{12} cm⁻²); only a minor change in effective lifetime is observed. We note that the TRCL curve is close to the TRPL curve for the higher injection condition; this is in-line with our matching initial carrier density estimates on the order of 10^{12} cm⁻² for TRCL (see Appendix D). (b) The TRPL decay curve from the $5 \mu\text{J cm}^{-2}$ condition is compared to an ideal AB model with varying IQE (black dashed lines). Only predictions with extremely low IQE can describe the TRPL data, but this is incompatible with the known high IQE of this sample (estimated at 64% [21,27]). The IQE steps between the AB curves are set to around 25%.

directly proportional to the modeled total number of carriers, $n_{\text{tot}}(t)$. It also allows us to equate luminescence-decay lifetimes away from the PD directly to τ_{bg} , as we do in Appendix D. Here, we justify this treatment.

In Fig. 9(a), we present decay curves for the studied sample at room temperature from both TRPL and TRCL away from PDs. Fitting the entire curves with Eq. (B1) reveals that all display near-perfect monoexponential decay over many decades in luminescence intensity. This observation already justifies treating τ_{bg} as carrier-density independent. Furthermore, since we know the sample is dominated by the radiative recombination rate in this carrier density range [21,27], it already implies that τ_r is near-constant across great changes in the carrier density.

This conclusion is reinforced by inspecting two TRPL decays gained at excitation densities, and hence initial carrier densities, which differ by over a factor of 70. The fits show that the effective lifetime, τ_{eff} , only decreases by approximately 35% for this near two-order-of-magnitude increase in the carrier density. Such behavior is incompatible with the ABC model prediction of $\tau_r \propto n^{-1}$. In fact, if we assume τ_r should have a density-dependence proportional to $n^{-\gamma}$, these two curves suggest the exponent is as minor as $\gamma \approx 0.1$. This leads to at most a 10% discrepancy between the true carrier lifetime and luminescence-derived lifetimes. Indeed, it has been demonstrated that the B

coefficient in the ABC model has an underlying dependence on the carrier density which increasingly suppresses the proposed $\propto n^2$ radiative rate closer to a linear relation as InGaN/GaN QWs become thinner [36]; for a 2.5 nm wide well, the corresponding τ_r dependence is already closer to $n^{-0.5}$ than n^{-1} . Consequently, carrier-density-independent τ_r for our extremely thin QWs (around 0.8 nm) is not a surprising observation.

It may be argued that this monoexponential decay behavior is rather due to a dominant SRH A term in a standard ABC model. However, this claim would be contradictory to this sample's high IQE, which was estimated to exceed 60% in Refs. [21,27]. To elucidate this contradiction, we compare one of the sample's decay curves to results from an ideal AB model as the underlying IQE in the model is decreased.

An AB model treats the carrier recombination rate as $dn/dt = An + Bn^2$, with the radiative component as Bn^2 and the SRH component as An . At any given time, the intensity collected in a luminescence setup is given by $I \propto Bn^2$ where the collection and extraction efficiency determine the proportionality constant. Solving analytically for I in terms of t while setting $n(t=0) = n_0$, we find

$$I(t) \propto \frac{A^2 B n_0^2}{(A e^{At} + B n_0 e^{At} - B n_0)^2}, \quad (\text{G1})$$

which gives τ_{eff} from a linear approximation of $\ln(I)$ at $t = 0$ as

$$\tau_{\text{eff}} = (2A + 2B n_0)^{-1}. \quad (\text{G2})$$

Meanwhile, the initial IQE is expressed as the ratio between the radiative and total recombination rates:

$$\eta_{\text{int}}(t=0) = \frac{B n_0}{A + B n_0}. \quad (\text{G3})$$

Combining Eqs. (G2) and (G3):

$$B = \frac{\eta_{\text{int}}(t=0)}{2\tau_{\text{eff}} n_0}, \quad A = \frac{1 - \eta_{\text{int}}(t=0)}{2\tau_{\text{eff}}}. \quad (\text{G4})$$

Substituting these expressions for A and B in Eq. (G1) gives the AB model $I(t)$ in terms of $\eta_{\text{int}}(t=0)$, τ_{eff} , and n_0 . We fix n_0 to 10^{12} cm^{-2} in agreement with our expected initial carrier density at an excitation fluence of $5 \mu\text{J cm}^{-2}$, and τ_{eff} to 3.7 ns to ensure the model will match the early decay behavior. This allows us to compute $I(t)$ for assumed initial IQE values, convoluting the result with the instrument-response function of our detector for a more accurate comparison with experimental data.

The resulting AB curves from an IQE of near 100% down to only 0.5% are displayed in Fig. 9(b). It is apparent

that the QW luminescence can only be described with the AB model if the IQE is far below 10%, yet we know that this is not the case; hence, we conclude that the radiative rate in this QW cannot be proportional to Bn^2 . This incompatibility firmly establishes a minor carrier-density-dependence in the radiative lifetime of our QW samples, thereby justifying our treatment in the main text and lending further confidence to the results from TRCL modeling.

The near-constant radiative lifetime observed in our sample likely results from a high excitonic fraction, which leads to predominantly monomolecular radiative recombination and consequently a radiative lifetime that is independent of carrier density. In fact, high excitonic fractions are expected for such thin InGaN QWs: recent modeling of $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}$ QWs accounting for alloy disorder revealed that the exciton binding energy increases from around 16 meV at a well width of 4 nm up to 47 meV at a width of 2 nm [54]. Extrapolating this trend conservatively to our 0.8 nm $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}$ QW yields an estimated exciton binding energy of around 70 meV. Applying Saha's equation at room temperature using this binding energy estimates an excitonic fraction in our QW exceeding 50% for the carrier densities generated in our experiments (on the order of 10^{19} cm^{-3} , see Appendix D).

The possibility of a high excitonic fraction in our sample raises the question of how the model presented in the main text would translate to an excitonic, rather than free electron-hole, interpretation. Since the model describes single-particle diffusion, this component can be applied equally well to excitons. However, the capture and recombination mechanisms for an exciton at a PD differ from those for individual electrons or holes.

In SRH recombination, an electron and a hole are captured sequentially, suggesting that an exciton would need to dissociate before it could undergo nonradiative capture at a defect. In addition, the Sommerfeld factor describing the Coulomb interaction between a charged defect and an electron or hole does not apply to a neutral exciton. In an excitonic picture, the PD delay time derived in the main text could therefore include a contribution from exciton dissociation rather than the Sommerfeld factor.

Overall, we retained the free electron-hole framework in the main text because theoretical descriptions of exciton nonradiative recombination at PDs remain limited and are beyond the scope of this work.

APPENDIX H: FULL ANALYSIS OF MODEL WITHOUT THE DELAY TIME

In the study, we claim that our initial diffusion model without any PD delay time fails to fit the single PD TRCL dataset in Fig. 2(e) of the main text. Here, we provide conclusive evidence for this claim.

Figure 10 compares the TRCL data to the initial model results for a variety of σ and $D_{||}$ values (the two free fitting

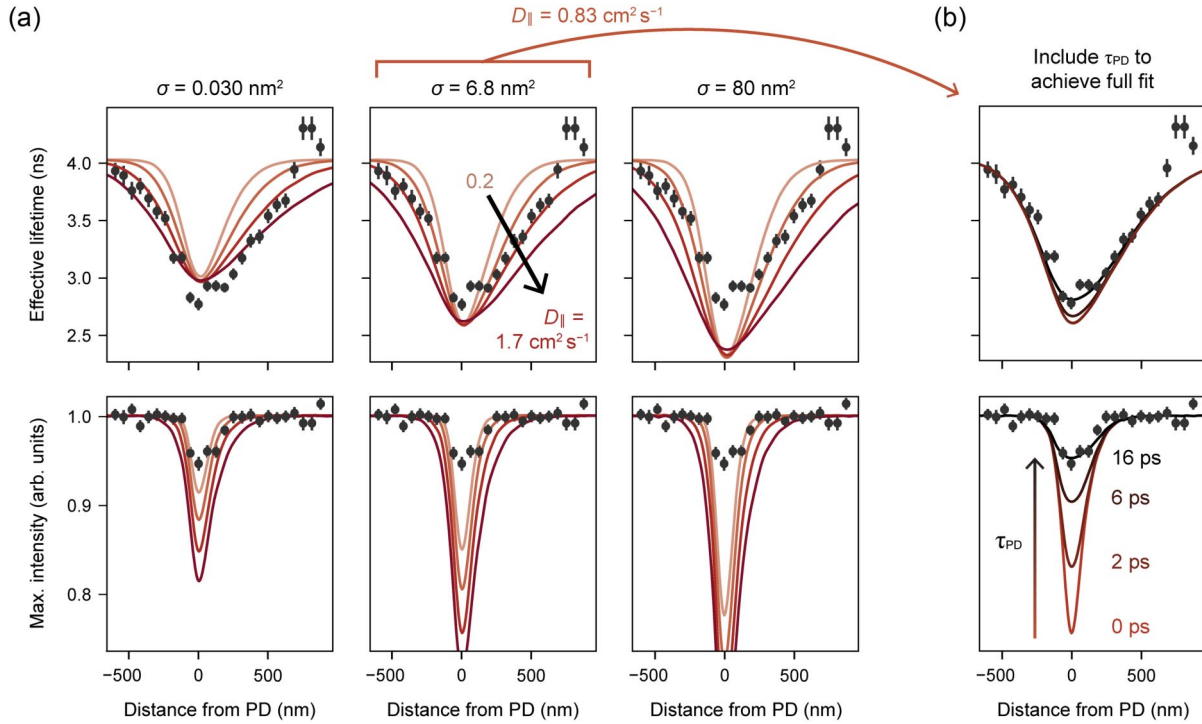


FIG. 10. (a) Comparison of TRCL data in Fig. 2(e) of the main text to the initial diffusion model without τ_{PD} , using average effective lifetimes and maximum intensities gained from applying Eq. (B1) to visualize the full 2D dataset. The effective lifetime behavior is only matched for a specific pair of σ and $D_{||}$ values; this same pair cannot model the maximum intensity. This deadlock can only be resolved by including τ_{PD} as a fitting parameter (b).

parameters). The data and model predictions are illustrated in a condensed form by fitting each decay curve in Fig. 2(e) of the main text with Eq. (B1) up to the isolation limit of $t = 2 \text{ ns}$, providing average effective lifetimes and maximum intensities as a function of injection position. We emphasize that these average effective lifetimes are only a metric by which to track changes in the decay shapes, since behavior when injecting close to the PD is not monoexponential.

We see that the behavior of the effective lifetimes with varying injection distance from the isolated PD can only be matched for a specific pair of σ and $D_{||}$ (6.8 nm^2 and $0.83 \text{ cm}^2 \text{ s}^{-1}$). However, for this same set of values the initial model completely fails to match the maximum intensities. Consequently, there is no way for a model with only σ and $D_{||}$ as free fitting parameters to accurately describe the TRCL data.

Including τ_{PD} allows for an extra degree of freedom, which resolves the lifetime-intensity impasse, since τ_{PD} mainly influences the maximum intensity with only a minimal effect on the lifetime (see the right of Fig. 10). Overall, this inspection of the fitting mechanism underlines the need for a PD delay time to match our TRCL dataset. Finally, we note that the impact of τ_{PD} on the maximum intensity starts to saturate as τ_{PD} increases; this lends a slightly larger uncertainty to our estimated 16 ps value of τ_{PD} .

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