

RESEARCH ARTICLE

Exciton Self-Trapping Enables Broadband Emission in Two-Monolayer Colloidal CdSe Quantum Wells

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ABSTRACT

Achieving intrinsic broadband emission in colloidal quantum wells (CQWs) via self-trapped excitons (STEs) is highly desirable for solid-state lighting, yet remains challenging due to their rigid lattices and inherently weak exciton-phonon coupling. In this work, we have linked the broadband emission from two-monolayer CdSe CQWs with extreme quantum confinement to the radiative recombination of STEs. This observation unlocks efficient intrinsic STE emission without extrinsic dopants or stacking formations, characterized by an exceptionally broad bandwidth (354 meV centered at 548 nm), high quantum yield (~90%), and shallow trapping barriers (~14.8 meV) enabled by strong exciton-phonon coupling (Huang-Rhys factor ~36.7). Ultrafast spectroscopic studies confirm rapid exciton self-trapping (~170 fs) and negligible exciton-exciton annihilation, reflecting the localized STE state. These atomically thin CdSe CQWs thus provide a chemically simple, highly efficient, and stable foundation for next-generation white-light emitters.

1 | Introduction

Two-dimensional (2D) colloidal cadmium chalcogenide quantum wells (CQWs) exhibit remarkably narrow exciton emission compared to 0D colloidal quantum dots and 1D colloidal nanowires owing to their thickness control with atomic layer precision [1–5]. Coupled with their low-cost synthesis in large quantities, highly efficient photoluminescence and electroluminescence emission, and remarkable stability, colloidal CdX (X = S, Se, Te) CQWs have garnered considerable attention as compelling candidates for laser gain media [6–8] and light-emitting diodes [9–11] since the pioneering synthesis by Ithurria et al. in 2008 [1]. Beyond monochromatic optoelectronic applications, recent efforts have

focused on achieving multicolored or broadband emission in CdX CQWs, driven by the technical demand for single-component white-light emitters in solid-state lighting: a critical advantage over traditional multicomponent phosphors [12]. For instance, incorporation of impurity transition-metal ions (e.g., Cu²⁺, Ag⁺, Mn²⁺ dopants) [13–15] and the rational design of lateral heterostructures [16, 17] have demonstrated the broadband bicolor emission in CdX CQWs.

Besides the above-mentioned extrinsic strategies, radiative recombination from self-trapped excitons (STEs) that are localized by surrounding lattices provides a feasible route to achieve intrinsic broadband emissions, as widely observed

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in low-dimensional lead halide perovskites (LHPs) [18–20]. Unfortunately, in comparison to LHPs with soft and ionic lattices, CdX CQWs have a rigid lattice with limited fluctuations upon photoexcitation [21, 22]. Both a weak temperature dependence of the excitonic emission linewidth and the sharpness of absorption/emission spectra indicate that the exciton-phonon coupling (EPC) involved in the heavy-hole transition is relatively weak (e.g., the EPC strength and the Huang-Rhys factor in 4-monolayer CdSe CQWs are ~ 6.6 meV and ~ 0.1 , respectively) [23, 24], which greatly hinders the observation of room-temperature STE emission. Recently, softening the rigid lattice by pushing CdX CQWs into the atom-cluster regime [25] and self-assembling of CdX CQWs into the stacking structure [26] have been proposed for the realization of STE, which, however, have introduced structural complexity and stability trade-offs.

In principle, pushing the quantum confinement to the limit could provide a desirable solution since the interaction strength between excitons and phonons scales inversely with the vertical thickness in 2D systems [21, 27, 28]. Herein, we have synthesized two-monolayer (2 ML) thick CdSe CQWs, which are the thinnest thermodynamically stable CQWs [3], to realize and investigate STEs. Steady-state photoluminescence (PL) spectroscopy has revealed that these ultrathin 2 ML CdSe CQWs show strong EPC with a Huang-Rhys factor of ~ 36.7 and a shallow self-trapping barrier of ~ 14.8 meV, which enables an efficient and broadband STE emission (548 nm, 354 meV) with quantum yields up to 90%. Using transient absorption spectroscopy techniques, we have uncovered that photoexcited excitons in 2 ML CdSe CQWs rapidly localize to intrinsic STEs within an ultrafast timescale of ~ 170 fs and an extremely small exciton-exciton annihilation rate ($\sim 7.3 \times 10^{-12}$ cm³/s), which is consistent with their highly localized nature. Our results demonstrate that the 2ML CdSe CQWs have the potential to pave the way for the rational design of high-efficiency white-light-emitting materials.

2 | Results

By using short-chain ligands with optimized reaction time and temperature to suppress vertical growth (see Methods), 2 monolayer (ML) CdSe CQWs, where the 2 Se crystalline planes are bound to 3 planes of Cd, as schematically shown in Figure 1a, were synthesized and dispersed in hexane [29]. The lateral size of the CdSe NPLs can be modified by changing the growth time, and these ultrathin 2 ML CdSe NPLs are rolled up to form tubular forms as their lateral area gets larger [29]. A transmission electron microscopy (TEM) image of rolled NPLs tubes having an average length of ~ 50 nm is shown in Figure 1b. The shape of the synthesized 2 ML CdSe CQWs is nearly ellipsoid with the lateral size of (62 ± 7) nm $\times$ (45 ± 6) nm as can be deduced from a few unrolled NPLs in the TEM image. Photoexcited properties of 2 ML CdSe CQWs are first characterized by the steady-state absorption and photoluminescence (PL) spectra at room temperature. As shown in Figure 1c, two discrete absorption peaks at ~ 372 and ~ 393 nm can be identified, which are consistent with the transition energy predicted by the potential well model with 2 ML vertical thickness [30] and thus, can be attributed to the electron light-hole (LH) transition and electron heavy-hole (HH) transition, respectively.

Unlike other CdSe CQWs with a sharp emission profile [1, 3], the PL spectrum of 2 ML CdSe CQWs is dominated by a broad emission band ranging from ~ 430 to ~ 720 nm with a PL quantum yield (PLQY) of $\sim 90\%$, which suggests the suppressed non-radiative centers. Generally, the radiative recombination from either STEs (i.e., excitons trapped in distorted lattices with strong electron-phonon couplings) [18, 20] or traps (i.e., when reducing the vertical thickness to a subnanometer level, surface defects could become unignored) [31–34] is expected to produce such a broad and featureless emission with a huge Stokes shift. More importantly, the PL profile of 2 ML CdSe CQWs is nearly independent of the excitation wavelength (Figure S1), which implies that it should come from a single excited state instead of different trap state transitions [20, 35]. Meanwhile, trap states will lead to an increase in the absorption just below the bandgap (i.e., sub-bandgap absorptions) in semiconductor materials [36, 37]. To test this, we have analyzed the second derivative of the absorption spectrum (the blue curve in Figure 1c), which is sensitive to identifying the weak and inconspicuous absorption feature [38]. These observations in Figure 1 support that the broadband emission in 2 ML CdSe CQWs is more likely to originate from STEs.

The nature of broadband emissions in 2 ML CdSe CQWs is first examined by checking the steady-state excitation power density-dependent PL at room temperature, which is shown in Figure 2a and Figure S2. With the excitation density ranging from 50 to 450 W/cm², the integrated intensity of this broadband emission in Figure 2b exhibits a linear dependence in the log-log plot with a power-law exponent of nearly one, suggesting that the radiative recombination comes from bound electron-hole pairs (excitons) in 2 ML CdSe CQWs [20, 39]. Please note that high-density interactions may allow a minor fraction of band-edge excitons to radiatively recombine before trapping occurs, which results in a weak free excitonic emission at high excitation intensities in Figure 2a. Time-resolved PL (TRPL) decay kinetics at different carrier densities (N_0) probed at ~ 535 nm are also presented in Figure 2c and Figure 2d to identify the origin of emitting species in 2 ML CdSe CQWs. In principle, the initial TRPL intensity at $t = 0$ (PL_0) after photoexcitation before recombination, which is proportional to the radiative recombination rate, should be proportional to N_0 for bound electron-hole pairs and N_0^2 for unbound electron and hole charges [6, 40]. As shown in Figure 2c and Figure 2d, PL_0 remains linear and does not show any saturation trend with N_0 increasing from $\sim 1.2 \times 10^{18}$ to $\sim 3.6 \times 10^{18}$ cm⁻³. This indicates that the broad PL emission in 2 ML CdSe CQWs originates from intrinsic excitons instead of electron/hole charges separated by extrinsic defect/trap states, which otherwise will show sublinear behavior and be saturated with this high carrier density. By fitting TRPL kinetics in Figure 2c with the exciton-exciton annihilation (EEA) model (i.e., $dN/dt = -N/\tau - \gamma N^2$, where τ is the exciton radiative recombination lifetime and γ is the EEA rate) [41, 42], γ is determined to be $\sim 7.35 \times 10^{-12}$ cm³/s, which is one or two orders of magnitude smaller than the value of free excitons (e.g., 10^{-10} to 10^{-11} cm³/s) reported in CdSe CQWs [43], implying a highly localized nature of excitons (i.e., STEs) in 2 ML CdSe CQWs.

We have further carried out the temperature-dependent PL measurements to investigate the strength of exciton-phonon coupling (EPC) for STE formation. As shown in Figure 2e and Figure

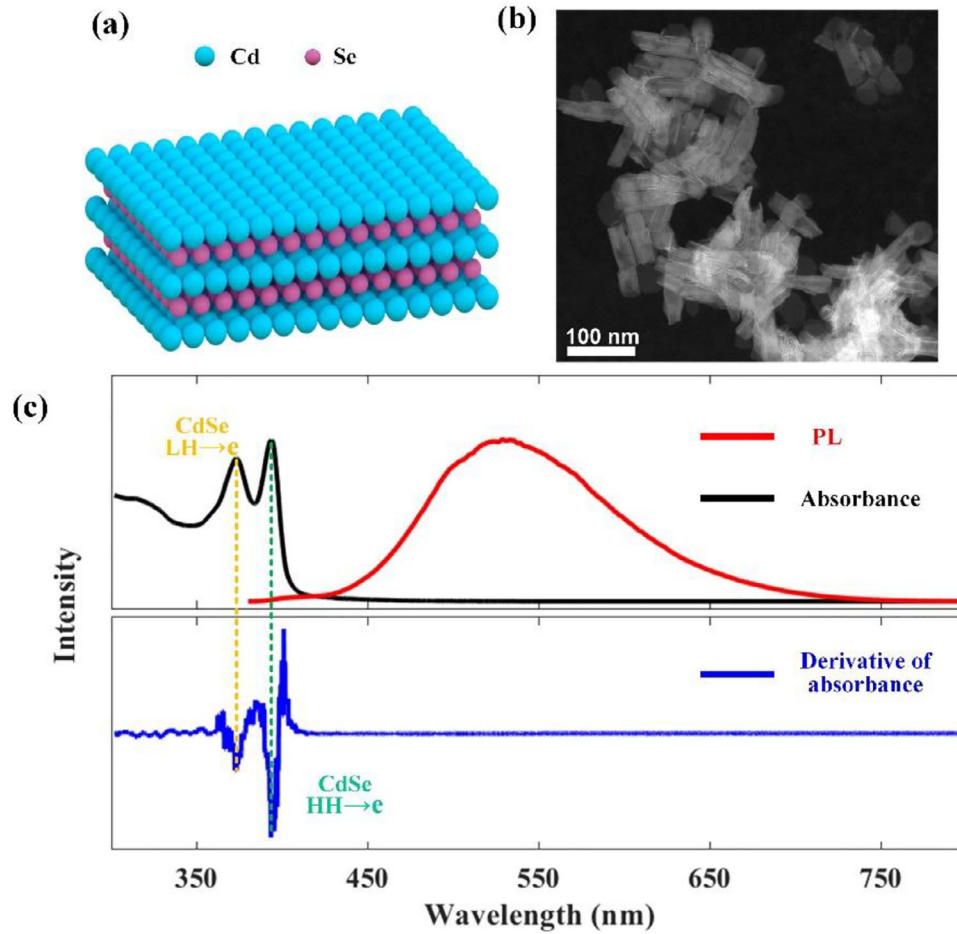


FIGURE 1 | Basic characterizations of 2 ML CdSe CQWs. (a) A schematic illustration of 2 ML CdSe CQWs. (b) TEM images of 2 ML CdSe CQWs. (c) Upper: Absorption (black) and photoluminescence (red) spectra of 2 ML CdSe CQWs. Lower: Absorption second derivative (blue) spectra of 2 ML CdSe CQWs.

S3, the intensities of this broadband emission decrease gradually with increasing temperature from 77 to 298 K, accompanied by the emission profile broadening, as commonly observed in 2D perovskites with STEs [44, 45]. Accordingly, the Huang–Rhys factor (S), which describes how strongly excitons are coupled to phonons, can be estimated based on the relationship between PL full-width-at-half-maxima (FWHM) and temperature using the Fröhlich longitudinal optical (LO) phonon broadening model [24, 44, 45]:

$$FWHM(T) = 2.36\sqrt{SE_{\text{phonon}}}\sqrt{\coth(E_{\text{phonon}}/2k_B T)} \quad (1)$$

where E_{phonon} is the LO phonon energy, and k_B is the Boltzmann constant. Based on the fitting presented in Figure 2f, the fitted phonon energy of ~ 25.2 meV is close to the value of the A_{1g} LO phonon mode in CdSe CQWs [46]. The Huang–Rhys factor is determined to be ~ 36.7 , which is comparable to the value reported for CdSe nanocluster (23.0) [47], stackings of CdSe CQWs (22.7) [26], $\text{Cs}_2\text{AgInCl}_6$ (38.7) [18], and Sb_2S_3 (38.5) [48], in which the STE is also observed. In contrast, the S value of 4 ML CdSe CQWs dispersed in solution is around 1 [20], and such a large S value in 2 ML CdSe CQWs indicates the strong EPC process, which benefits from pushing the vertical quantum confinement to the limit and accounts for the formation of STEs with the broadband emission.

Moreover, the Arrhenius plot of the integrated PL intensity versus the temperature is shown in Figure S4 to quantify the energy barrier for STE formations, which yields a small energy barrier of ~ 14.8 meV, agreeing well with the dominant broadband emission from STEs.

To follow the exciton self-trapping process in real time, we have conducted femtosecond transient absorption spectroscopy (fs-TA), in which 2 ML CdSe CQWs are excited by a 310 nm, ~ 100 fs pump pulse with a carrier density of $\sim 1.4 \times 10^{19} \text{ cm}^{-3}$ (see Methods). The 2D pseudo-color plot of TA spectra is shown in Figure 3a and Figure S5. Besides the regular bleaching bands (i.e., the absorption change ΔA is negative) in Figure 3b, which are located at ~ 372 and ~ 393 nm corresponding to LH and HH excitonic transitions, we have observed a broad below-bandgap photoinduced absorption feature (i.e., $\Delta A > 0$ as indicated by the dashed black box), whose energy is in agreement with the observed below-bandgap broadband emission. This observation further excludes the involvement of traps/defects since trapping of photogenerated charge carriers will block allowed transitions within the absorption tail and generate negative features in the TA spectrum [37, 49]. Considering that no free carriers are expected in this ultrastrong quantum confinement, we attribute the photoinduced absorption band to the formation of STE states in the below-gap region following photoexcitations, which has also been

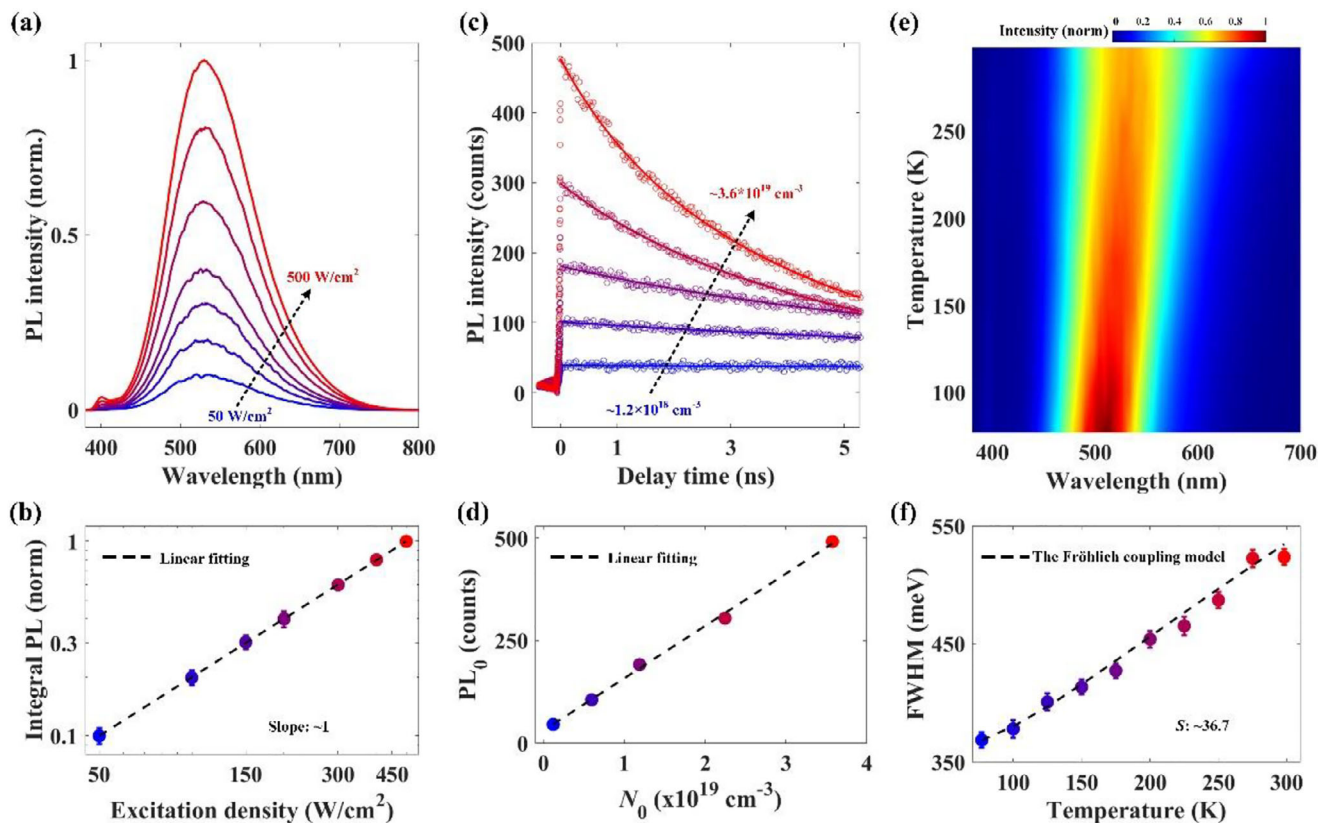


FIGURE 2 | PL properties of 2 ML CdSe CQWs. (a) PL spectra under 365 nm excitation light with different intensities. (b) Power-dependent PL intensity and its linear fitting. (c) trPL decay kinetics with different photoexcitation densities. (d) Initial trPL intensity PL_0 as a function of N_0 and its linear fitting. (e) Contour plots of the temperature-dependent PL emission spectra excited at 365 nm. (f) Change in PL line width with temperature. The black curve is a fitting according to the configuration model.

viewed as the direct evidence of STEs in perovskites [50, 51]. Meanwhile, the kinetics of this photoinduced absorption band are fitted based on a double-exponential function convoluted by the instrument response function. As shown in Figure 3c, the STE process in 2 ML CdSe CQWs exhibits a rapid turn-on time (~ 170 fs), which coincides with the fast decay component (~ 213 fs) of HH excitons in Figure S6, indicating that the decay of free excitons at the band-edge leads to STE formations in 2 ML CdSe CQWs with a shallow potential barrier. Note that this STE formation time is comparable to the oscillation period of the A_{1g} LO phonon (i.e., ~ 25.2 meV or ~ 203.3 cm $^{-1}$ corresponds to an oscillation period of ~ 164 fs), confirming the critical role of EPC in exciton self-trapping. Such an ultrafast STE formation time indicates the diffusion length of excitons on the order of sub-1 nm, which implies that photoexcited excitons are captured by the intrinsic lattice distortion (self-trapping) long before they have time to diffuse to extrinsic defects. Furthermore, we have compared the STE dynamics probed at different wavelengths, which display a nearly identical rise time in Figure 3d and Figure S7, suggesting again that the photoinduced absorption band originates from a single excited STE state instead of different trap state transitions [52]. Exponential fitting of STE decays reveals a fast component of ~ 0.88 ps associated with the nonradiative relaxation of STEs and slow components related to the radiative recombination of STEs.

To obtain further clues about the self-trapping process of excitons in 2 ML CdSe CQWs, excitation fluence-dependent fs-TA

measurements have been conducted with the carrier density ranging from 1.1×10^{18} to 4.1×10^{19} cm $^{-3}$ (2D color plot of the TA spectra with different carrier densities is shown in Figure S8). Similar to the analysis in Figure 3c, we have fitted the initial kinetics of PIA signals to extract the turn-on lifetime of STEs with different carrier densities (Figure 4a; Figure S9). It is well-established that STEs can form either by direct lattice capturing of photoexcited excitons or through a two-step mechanism: the pretrapping of electrons or holes first occurs, and then STE forms via a bimolecular process [20, 48]. These two mechanisms can be differentiated by checking the trapping dynamics as a function of photoexcited exciton density (i.e., the first monomolecular trapping process is independent of N_0 while the second bimolecular trapping process will be influenced by the number of pretrapped and remaining carriers). The STE formation time as a function of N_0 is plotted in Figure 4b, which shows a nearly unchanged value of ~ 170 fs. This negligible carrier density dependence confirms the spontaneous and geminate localization of free excitons to STE states with a negligible energy barrier. In addition, the N_0 -dependent amplitude of PIA signals at ~ 420 nm exhibits a uniform linear growth trend (Figure 4c; Figure S10), correlated with the PLs result shown in Figure 2. More importantly, the amplitude of PIA signals does not show any saturation signature even at 4.1×10^{19} cm $^{-3}$, which further excludes the contribution of extrinsic defects considering that the trap densities [34, 53] are in the range of $\sim 10^{12}$ to 10^{16} cm $^{-3}$ in the colloidal nanocrystals with a volume of 8000 nm ^{3} . Hence, the mechanism of the

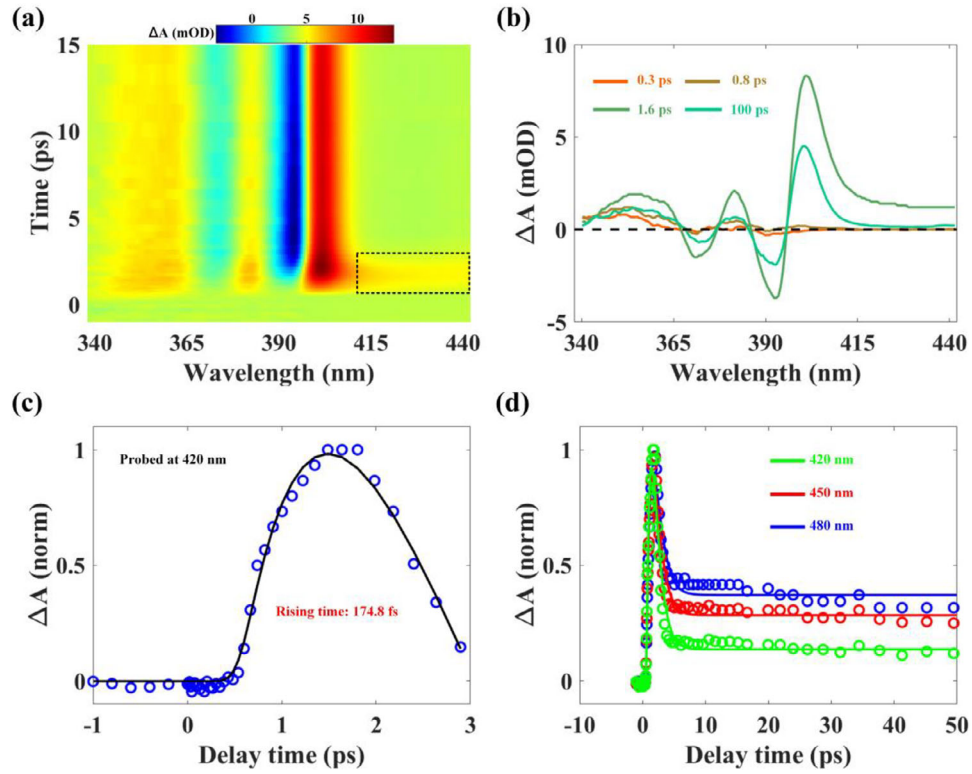


FIGURE 3 | Femtosecond transient absorption study of 2 ML CdSe CQWs with $N_0 = \sim 1.4 \times 10^{19} \text{ cm}^{-3}$. (a) 2D color plot of the TA spectra excited with the 320 nm laser pulse. The dashed black box indicates the PIA signal induced by STEs. (b) Representative TA spectra cuts at different delay times. (c) TA kinetics probed at 420 nm (blue dots) with the exponential fitting (the black line). (d) TA kinetics probed at different wavelengths (dots) with exponential fittings (solid lines).

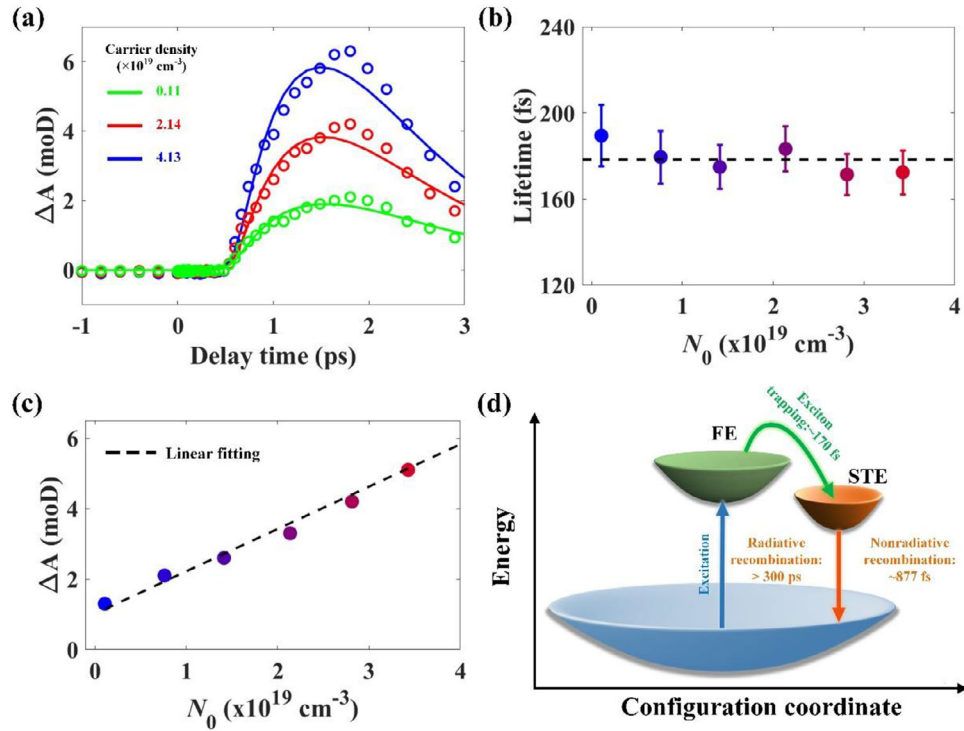


FIGURE 4 | Carrier density-dependent STE dynamics in 2 ML CdSe CQWs. (a) TA kinetics probed at 420 nm (dots) with different carrier densities. The solid lines are the exponential fittings. (b) The STE formation time as a function of carrier densities. (c) Peak amplitude of TA kinetics probed at 420 nm with different carrier densities. The dashed line is a linear fit. (d) Schematic illustration of the STE formation and recombination dynamics in 2 ML CdSe CQWs.

ultrabroadband emission due to STEs [25, 54] can be depicted in Figure 4d. Upon photoexcitation, 2 ML CdSe CQWs are excited to the free exciton (FE) excited state. The photogenerated FEs quickly form the STE state induced by strong exciton-phonon couplings with a time scale around 170 fs. Then, the STE state relaxes to the ground state via either the fast nonradiative process (with a time scale of ~ 0.9 ps) or the long-lived emissive process with a lifetime at the nanosecond time scale.

3 | Conclusion

In summary, this work establishes an intrinsic pathway toward high-performance broadband emission in CdX CQWs without compromising material stability or simplicity. By shrinking the thickness rather than introducing dopants or stacks, we have altered the exciton-phonon interaction landscape and promoted efficient self-trapping with minimal energetic barriers. The resulting near-unity quantum yield ($\sim 90\%$) and exceptionally broad STE emission (354 meV bandwidth) directly address the performance metrics required for single-component white-light sources. The observed ultrafast exciton localization dynamics (~ 170 fs) and suppressed exciton annihilation further confirm the formation of a highly localized STE state. These findings transform ultrathin CdX CQWs from purely narrow-band emitters into a compelling candidate for high-efficiency solid-state lighting technologies.

4 | Methods

4.1 | Material Synthesis

4.1.1 | Synthesis of 2 ML CdSe NPLs

860 mg of cadmium acetate dihydrate, 1 mL of oleic acid, and 50 mL of octadecene were placed into a three-neck flask and degassed under vacuum for an hour at room temperature. Then, the mixture was heated to 100°C under nitrogen flow, and 2 mL 1 M Se dissolved in trioctylphosphine (TOP-Se) was injected swiftly. The mixture was kept at 100°C 12 h for the growth of 2 ML CdSe NPLs. After that, the solution was cooled down to room temperature and precipitated by centrifugation after adding ethanol. The precipitated NPLs were dispersed and kept in hexane.

4.1.2 | Transient Absorption Measurements

A 1 kHz regenerative amplifier with an 800 nm output wavelength and a 100 fs pulse width, which is seeded by the 80 MHz mode-locked Ti-sapphire oscillators, was used to produce the pump pulse with a maximum energy of mJ. The pump pulse then went through the optical parametric amplifier (OPA) to produce the desired excitation energy for fs-TA experiments. To generate the white light continuum probe beam spanning from 400 to 1 500 nm, we have split a small portion of the fundamental 800 nm pulse from the regenerative amplifier with an energy of ~ 10 μJ and then incident on sapphire crystals with different thicknesses for the continuum probe beam generation. A commercial spectrometer was used to collect the probe beam

spectra in the ultraviolet-visible region with and without the pump excitations.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

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

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.

Supporting File: lpor70967-sup-0001-SuppMat.docx.