

Electro-Optical Evaluation of Cu^{1+} and Cu^{2+} States in Copper-Doped CdSe Colloidal Quantum Wells

Junhong Yu,* Ke Wang, Yadong Han, Zhenzhong Lian, Songyan Hou, Chang Cao, Hilmi Volkan Demir, Jacek J. Jasieniak, and Manoj Sharma*



Cite This: *ACS Photonics* 2026, 13, 1193–1199



Read Online

ACCESS |



Metrics & More



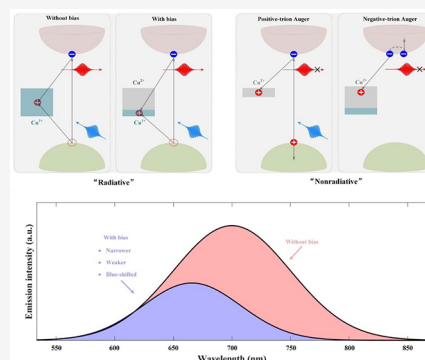
Article Recommendations



Supporting Information

ABSTRACT: Resolving the ambiguous oxidation state of copper dopants responsible for the large Stokes-shifted emission in CdSe colloidal quantum wells (CQWs) is critical for harnessing their emerging optoelectronic properties. Employing carrier injection to tune the population distribution between $\text{Cu}^{1+}/\text{Cu}^{2+}$ centers and in situ monitoring of the copper-related emission (CE), we have revealed that the CE band undergoes blueshifting, intensity quenching, and line width broadening with gradually increased Cu^{2+} states. Time-resolved CE dynamics and intragap absorption further confirm that Cu^{2+} states generate narrow, inefficient emission with minimal Stokes shifts due to trap-mediated Auger recombination, reduced radiative center energy spanning, and Fermi-level shifts. Accordingly, we identify Cu^{1+} as the dominant species that produces the bright, broad CE band with its hallmark large Stokes shift. This work not only presents mechanistic clarifications but also provides an effective approach to electrically modulate defect emissions in CQWs.

KEYWORDS: copper-doped, CdSe, colloidal quantum wells, colloidal nanoplatelets, spectroscopy



INTRODUCTION

Two-dimensional (2D) CdSe semiconductor colloidal quantum wells (CQWs) offer compelling advantages for optoelectronics, including low cost, giant oscillator strength, strong excitonic interactions, large absorption cross sections, and efficient narrowband emissions.^{1–4} These attributes have spurred rapid advances in a range of CQW applications, such as lasers,^{5,6} light-emitting diodes,^{7–12} and photodetectors.^{13–16} To further enhance CQW versatility, introducing transition metal ions (e.g., Mn, Cu, Ni, Ag) into the host lattice provides a distinct pathway, beyond heterostructure formation and wave function engineering, to tailor optical, electronic, and magnetic properties. Among these dopants, Cu has been extensively studied in CdSe CQWs^{17–23} for its ability to create atomic-like intragap states, improve emission efficiency, prolong radiative recombination lifetimes, and generate large Stokes-shifted copper-related emission (CE).

Despite impressive demonstrations of Cu-doped CdSe CQW in various devices, the origin of the CE band remains contentious. An ongoing debate is whether the emitting Cu defect exists in the +1 or +2 oxidation state.^{24–31} As shown in Figure 1a, both Cu^{1+} ($[\text{Ar}]3d^{10}$) and Cu^{2+} ($[\text{Ar}]3d^9$) states can support emission, albeit through distinct mechanisms. Cu^{2+} can act as an optically active acceptor with a 3d hole that can radiatively capture a conduction band electron. In contrast, Cu^{1+} has a filled 3d shell, and hole capture from the valence band (VB) is required to activate the CE process. Several studies^{32–38} have probed the oxidation state of emitting Cu

centers in CdSe nanocrystals, yet reported evidence remains contradictory (e.g., magneto-optical spectroscopy^{25,26} suggests Cu^{2+} , while X-ray absorption spectroscopy^{37,38} indicates Cu^{1+}). Therefore, resolving this oxidation-state ambiguity becomes a prerequisite to accurately understanding the optical, electronic, and magnetic properties of Cu dopants.

In this work, we have conducted comprehensive electro-optical spectroscopic investigations to gain further insights into the nature of the Cu oxidation state involved in the radiative recombination process. By electrically injecting external carriers into Cu-doped CdSe CQWs, which will convert Cu^{1+} to Cu^{2+} via hole capturing, we have identified the peak wavelength blueshifting, emission intensity suppression, and line width broadening of the CE band. Combined with the time-resolved CE dynamics and in situ steady-state intragap absorption spectra during carrier injections, our findings suggest that Cu^{2+} states render the dark and narrow dopant emission with small Stokes shifts, which stem from trap-assisted Auger recombination, reduced radiative-center energy spanning, and Fermi-level elevation. We further conclude that

Received: December 2, 2025

Revised: January 20, 2026

Accepted: January 22, 2026

Published: January 29, 2026



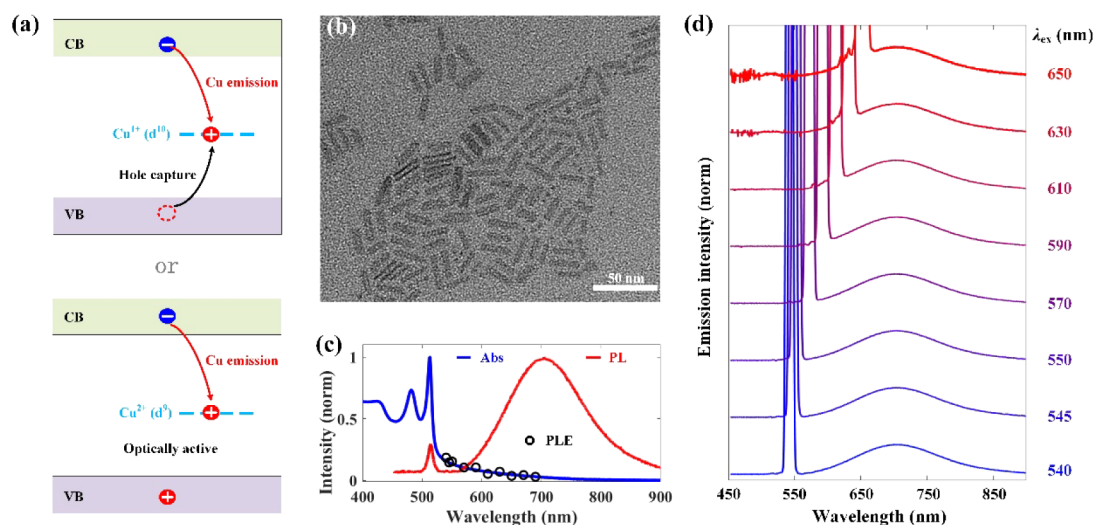


Figure 1. Optical properties of Cu-doped 4 ML CdSe CQWs. (a) Schematic illustrations of the different roles of Cu^{1+} (upper) and Cu^{2+} (lower) on the photoluminescence properties. (b) TEM image of Cu-doped 4 ML CdSe CQWs. The scale bar is 30 nm. (c) Optical absorption (blue curve) and PL spectra (red curve, excitation wavelength: 355 nm) of Cu-doped CQWs (~ 70 copper dopants per CQW) in toluene. Black squares represent the PLE spectrum of the copper emission probed at ~ 1.77 eV (~ 700 nm). (d) 2D copper emission (CE) spectra under progressively lower energy excitation from 540 to 650 nm. The saturated brown areas indicate the laser excitation spectra.

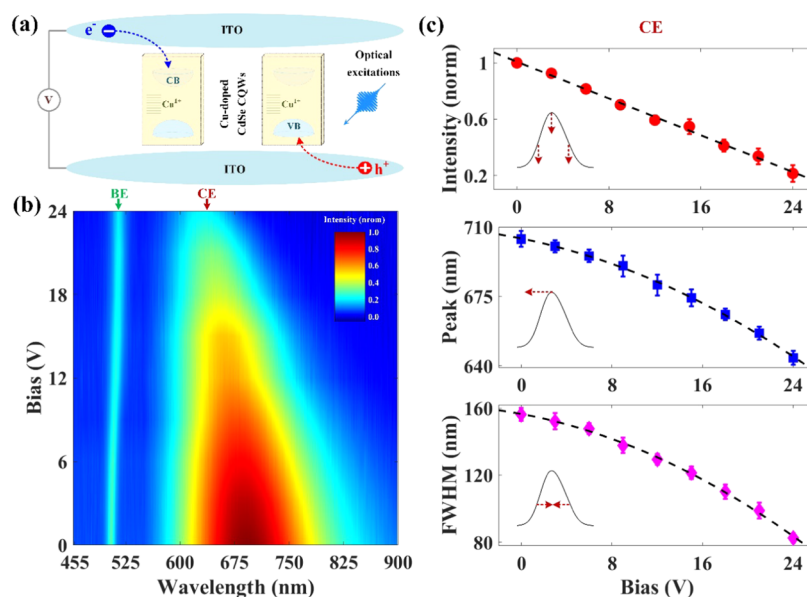


Figure 2. Behavior of copper emission under different applied biases. (a) Schematic illustration of the electric device configuration, in which Cu-doped CQWs are sandwiched between two ITO substrates. (b) 2D emission map of Cu-doped CQWs under different applied biases under 400 nm CW laser excitation. BE: the bandedge emission. CE: the copper emission. BE and CE are indicated by the green and red arrows, respectively. (c) Bias-dependent intensity (the upper panel), peak wavelength (the middle panel), and the fwhm (the lower panel) of CE.

Cu^{1+} states are the origin of the characteristically bright, broad CE band with large Stokes shifts in Cu-doped CdSe CQWs.

RESULTS

Four-monolayer (ML) Cu-doped CdSe CQWs with a quantum yield of $\sim 80\%$ were synthesized by using an optimized recipe (see **Methods**),^{39,40} which achieves high aspect ratios to preclude face-to-face stacking in the solid state (see the TEM image in **Figure 1b**). **Figure 1c** shows the characteristic absorption and emission spectra of a dilute toluene solution of Cu-doped CQWs with an average of 70 dopants per CQW. Upon incorporation of copper, an intense, broad CE band peaking at ~ 700 nm was observed with a huge

bandwidth of ~ 180 nm and an ultralong lifetime of ~ 457 ns (see **Figure S1**). Here it is found that the CE band is red-shifted relative to the electron/heavy-hole (HH) transition at ~ 508 nm, and the weak residual excitonic emission is peaked at about ~ 512 nm. Although the copper dopants do not change the above bandgap absorption feature (i.e., the electron/heavy-hole and electron/light-hole transitions) in CQW hosts, the intragap absorption properties could shed light on the oxidation state of Cu centers.^{41–43} Specifically, the Cu^{2+} state is absorption passive because promotion of its 3d electron into the conduction band would create a forbidden 3d⁸ final state; accordingly, its presence does not perturb the absorption edge.

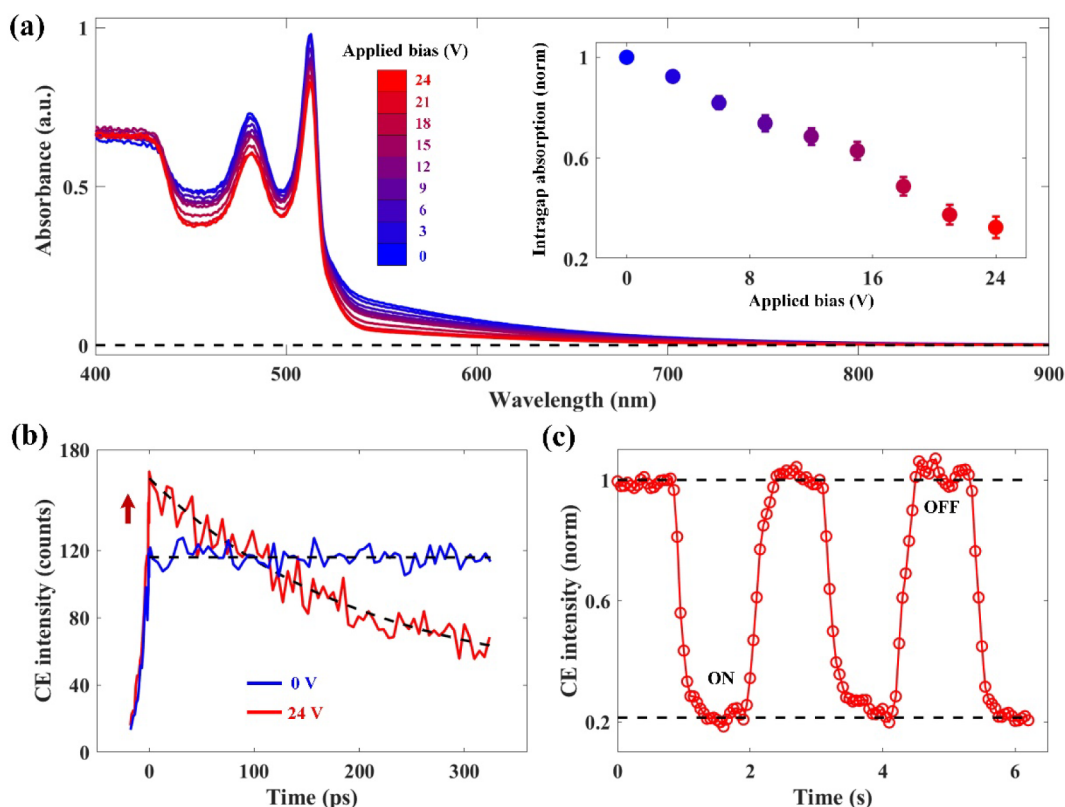


Figure 3. Steady-state intragap absorption spectra and time-resolved CE dynamics during carrier injections. (a) Absorption spectra of Cu-doped CdSe CQWs under different biases. The inset is the normalized integrated area of the copper term as a function of the applied biases (see fitting processes using the quantum-well absorption model in [Supplementary Note 1](#)). (b) The ultrafast CE dynamics without the bias (the blue curve) and with the bias of 24 V (the red curve) following the excitation by a 400 nm laser pulse. The black dashed line is the exponential fitting. (c) CE intensity under 400 nm CW laser excitation modulated by a periodic bias at a frequency of 0.5 Hz.

In contrast, the Cu^{1+} state is absorption active as one of its 3d electrons can be optically excited into the CB by leaving the copper cation in the $3d^9$ configuration. The Cu^{1+} to CB transition is therefore expected to broaden the absorption edge, which is consistent with our observation in [Figure 1c](#). This sub-bandgap Cu^{1+} to CB transition is also supported by the subgap excitation emission spectra. As shown in [Figure 1d](#), the copper emission can still be observed in the continuous-wave excitation with progressively lower excitation energy below the heavy hole exciton transition energy of the CdSe CQW host (i.e., tuning the excitation wavelength from 540 to 650 nm). This observation further validates that 3d electrons of Cu^{1+} can be promoted to the CQW CB by direct optical excitation, following the reaction $[\text{Ar}]3d^{10} + h\nu \rightarrow [\text{Ar}]3d^9 + e_{\text{CB}}$, then the promoted electron in the CB can radiatively recombine with the temporarily photoconverted Cu^{2+} . Consequently, the PLE excitation (PLE) spectrum collected at the CE maximum overlays nearly perfectly the intragap absorption tail (the black circles in [Figure 1c](#)), while the CE profiles remain identical irrespective of whether excitation occurs above or within the tail.^{44,45} These observations constitute the first experimental evidence that the broad CE is more likely to originate from the Cu^{1+} center.

Independent confirmation of the above proposed mechanism is provided by electro-optical spectroscopy, which in situ monitors the CE properties during carrier injection.⁴⁶ As shown in [Figure 2a](#), we have employed a simple device structure by sandwiching a Cu-doped CQW layer with two ITO electrodes. Based on the energy levels of ITOs (−4.8 eV)

and the CB minimum (−3.41 eV), the VB maximum (−5.82 eV) of 4 ML CdSe CQWs, external electrons and holes will be injected into the CB and VB of Cu-doped CQWs, respectively. The carrier injections establish distinct quasi-Fermi levels for electrons ($E_{\text{F},e}$) and holes ($E_{\text{F},h}$). The probability of a Cu^{1+} center capturing a hole (i.e., being oxidized to Cu^{2+}) is governed by the energy alignment between the $\text{Cu}^{2+}/\text{Cu}^{1+}$ state and $E_{\text{F},h}$. As the injection current increases, the hole quasi-Fermi level shifts downward toward (or below) the $\text{Cu}^{1+}/\text{Cu}^{2+}$ charge-transition level, increasing the hole concentration. This raises the Fermi–Dirac distribution function for holes, thereby significantly increasing the rate of hole capture by the Cu^{1+} states and raising the population ratio between $\text{Cu}^{2+}/\text{Cu}^{1+}$. It is worth mentioning that the current density flowing through the entire device is less than 40 mA/cm² with the CQW thickness of ~400 nm (see [Figure S2](#)), indicating minimal heating effect during the current injection process.⁴⁷ [Figure 2b](#) presents the 2D colormap of the emission spectra of Cu-doped CQWs with different applied biases ranging from 0 to 24 V (also see the complete set of emission spectra in [Figure S3](#)). We follow the evolution of the three main characteristics of the CE band as a function of the applied bias in [Figure 2c](#): the intensity (the upper panel), position of the maximum (the middle panel), and the emission bandwidth (the lower panel), which show a clear correlation between the characteristic CE features and the applied bias.

With gradually increased carrier injection densities, the CE maximum blueshifts pronouncedly by ~70 nm, whereas the intensity drops dramatically by ~81% with an emission line

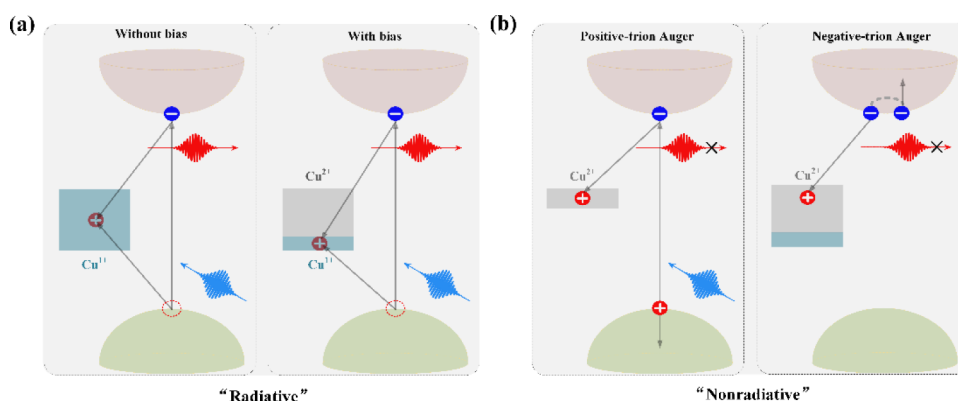


Figure 4. Recombination mechanism of Cu-doped 4 ML CdSe CQWs upon applying bias. (a) Schematic representation of the radiative recombination in Cu-doped CdSe CQWs without and with bias. Hole injections convert Cu^{1+} to Cu^{2+} , which results in a blue shift and narrowing of CE. (b) Schematic representation of the nonradiative recombination in Cu-doped CdSe CQWs. The trap-assisted Auger recombination process significantly reduces the intensity of CE.

width narrowing of ~ 80 nm. Here, the measured spectra represent an average over the entire device, and the nonuniformity of quasi-Fermi levels or carrier injections is not considered. When semiconductor nanocrystals are embedded in electronic devices, several mechanisms are expected to have an impact on the emission properties. Recently, Brovelli and coworkers have reported on the filling of intrinsic trap states (i.e., unpassivated surface and volume defects in the CQW host) with applied external potentials to change the Fermi level.²⁶ However, this explanation is incompatible with the CE trends observed in our experiments, since reducing the carrier trapping by intrinsic defects will only enhance the emission intensity and not affect the emission energy and line width. In addition to the charge injection, the electric field may also modulate the emission properties of CdSe CQWs through the Stark effect, as demonstrated by Woggon and coworkers.⁴⁸ To preclude contributions of the Stark effect, we have isolated the electrode/CQW contact by introducing thin layers of polyvinylpyrrolidone (PVP) between the CQW and the ITO layers.^{47,49} Direct charge injection flowing through such a device is almost prohibited (the maximum current is decreased to several μA in Figure S4). Correspondingly, CE emission shows negligible dependence on the applied biases up to 24 V (see Figure S5), confirming that the charge injection is the primary cause of CE modulations. Besides the conducted control experiment, future studies employing unipolar device architectures could further refine the role of carrier injections (e.g., using a semiconductor layer with selective band alignment to ensure only one type of carrier is injected).

Recalling the impact of different oxidation states on the absorption edge discussed in Figure 1c, the conversion of Cu^{1+} into Cu^{2+} can be further confirmed by monitoring the change of intragap absorption spectra during carrier injections. Based on the quantum well absorption model proposed by Elliott et al.,⁵⁰ which has been widely used in CdSe CQWs^{51,52} and Ruddlesden–Popper perovskites,⁵³ the absorption spectra of Cu-doped CQWs under different biases in Figure 3a can be decomposed into excitonic contributions and the continuum edge of Cu^{1+} intragap absorption (see Figure S6 for details). Accordingly, the population reduction of Cu^{1+} states can be estimated by integrating the decomposed intragap absorption band, provided that the Cu^{2+} state is absorption passive and does not perturb the absorption edge.^{42,44} A plot of the

integrated result as a function of the applied bias in Figure 3a indicates that the Cu^{1+} population at 24 V drops by over 74% compared to the value without applied bias, which is roughly consistent with the CE intensity reduction in Figure 2c and supports the critical role of Cu^{1+} states in dopant emissions. The suppression of the distinct intragap absorption feature, instead of a noticeable shift or broadening of the excitonic feature, has further ruled out the contribution of the Burstein–Moss effect. It is worth mentioning that without optical excitation, the Cu-doped CQWs with applied bias are nonemissive, although the majority of copper centers are in the +2 state due to the hole injection, and additional electrons are injected into the host CB. This implies that the three-particle trap-assisted Auger recombinations will open up (i.e., the recombination energy between the CB electron and the hole of the in-gap Cu^{2+} center is nonradiatively exciting another electron or hole to a deeper energy level).^{27,46}

This Auger recombination process can be validated by the time-resolved CE dynamics. As shown in Figure 3b, the recombination kinetics of CE become faster and the initial signal amplitude increases relative to the neutral case with the applied bias of 24 V. This is a characteristic signature for a three-particle (i.e., a trap state trion consist of a CB electron, a localized hole on a Cu^{2+} center, and an additional, third charge carrier) Auger process: the nonradiative Auger recombinations result in a short lifetime, and the doubled radiative decay channels in a three-particle scenario lead to an enhanced initial amplitude.^{27,49,54} For a cross-check, we have shown in Figure S7 that without carrier injection, such an Auger process is absent when only applying an electric field onto the Cu-doped CQW film without carrier injections. Analysis of the fast decay component in Figure 3b yields a time constant of ~ 208.7 ps for this Auger process, which matches the time constants when electrons trap rapidly to deep defect states near the VB edge in CdTe NCs via Auger excitation of photogenerated holes.⁵⁵ Moreover, the CE intensity under 400 nm cw laser excitation modulated by a periodic voltage of 24 V at a frequency of 0.5 Hz is presented in Figure 3c, which suggests that the carrier injection is a reversible process. We can determine that the response time of <100 ms for the CE change is much faster than the diffusion process of transition metal ions and the dissipation process of electric joule heating,^{47,56} which typically occur with time scales longer than seconds, implying again the dominant role of carrier injection on the CE modulation.

Based on the above experimental results, we schematically show in Figure 4a,b the postulated roles of Cu^{1+} and Cu^{2+} in the CE process. We suggest that the ground state of copper dopants in CdSe host is Cu^{1+} , which can be verified based on the strength of intragap absorption due to Cu^{1+} -to-CB transitions, the observation of CE under below-bandgap excitations, and the darkening/narrowing/blue-shifting trends of CE with carrier injections. Specifically, the applied bias will both inject holes and electrons into the VB and the CB of the CdSe host, respectively. The injected holes convert Cu^{1+} into Cu^{2+} and with the injected carriers, the Cu^{2+} states mainly behave as dark centers to quench the dopant luminescence due to the three-particle trap-assisted Auger recombination process: the Cu^{2+} center traps one CB electron and transfers the energy nonradiatively to another CB electron or a hole in the VB.^{27,46,54} Although with limited wave function overlaps, this trap-assisted Auger recombination could happen efficiently since one of the three carriers (i.e., the hole in Cu^{2+} states) is highly localized, which reduces the momentum conservation requirement, similar to previously reported trion Auger process in CdTe nanocrystals with deep volume defects⁵⁵ and Cu-doped CdSe/CdS nanocrystals with excess CB electrons.²⁷ Note that the three-particle Auger process with Cu^{2+} states occurs much faster compared to the radiative recombination of CE,^{15,57} which could reasonably explain the absence of CE in our device directly with applied bias. With the full consumption of the electric injected carriers via the three-particle Auger process and the recovery of Cu^{1+} state, optical excitations will again activate the CE process (i.e., the hole localization in Cu^{1+} states occurs with the time scale of 1–4 ps).^{22,45} Under these circumstances, CE with optical excitations will become blueshifted and narrower, due to a progressive reduction in the energy distribution of Cu^{1+} states and the larger energy separation between the CB minimum and Cu^{1+} states with external carrier injections.^{25,26}

CONCLUSION

In summary, our study definitively resolves the long-standing controversy regarding the oxidation state of copper dopants in CdSe colloidal quantum wells. By integrating in-operando spectral measurements with external carrier injection, we pinpoint Cu^{2+} as generating only spectrally narrow, inefficient photoluminescence due to detrimental electronic effects, contrasting sharply with the bright, broad-band photoluminescence attributable to Cu^+ . Concomitant time-resolved CE dynamics and intragap absorption measurements further reveal that the Cu^{2+} pathway is quenched by trap-assisted Auger recombination and a rigid Fermi-level rise. The identification of Cu^+ as the optically active center provides a universal mechanistic foundation for developing advanced transition metal-doped CQW emitters.

METHODS

Synthesis of High-Aspect-Ratio Cu-Doped CQWs

Four ML Cu-doped CdSe CQWs are synthesized by following our previously reported high-temperature nucleation doping method.^{32,36}

Electro-Optical Spectroscopic Investigations

The glass substrate with ITO is carefully cleaned to remove any organic residue. The Cu-doped CQWs in hexane (~15 mg/mL) are deposited on the ITO layer by the spin-casting technique. Before the drying of the CQW solution, two identical substrates with Cu-doped CQWs are stuck together to form the ITO/CQW/ITO sample. For

the control sample, the PVP dielectric layer is deposited on top of the ITO film by spin-casting (PVP dissolved in ethanol, 20 mg/mL, spin duration: 45 s, spin velocity: 2000 rpm). All the device fabrication processes are made inside the glovebox, and to remove any residual strains, we bake each device at 60 °C in a nitrogen environment for 20 min. A DC voltage power supply is used for carrier injections, and the current flowing through the device is recorded by the Keithley SourceMeter.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsphotonics.5c02900>.

PL decay dynamics of the BE and the CE; characterizations of the ITO/CQW/ITO and ITO/PVP/CQW/PVP/ITO samples; emission spectra of the ITO/CQW/ITO and ITO/PVP/CQW/PVP/ITO samples with different applied biases; analyzing the bias-dependent absorption spectra; ultrafast CE dynamics of the ITO/PVP/CQW/PVP/ITO sample (PDF)

AUTHOR INFORMATION

Corresponding Authors

Junhong Yu – College of Physics and Electronic Engineering, Chongqing Normal University, Chongqing 401331, China; orcid.org/0000-0001-6136-552X; Email: jyu012@e.ntu.edu.sg

Manoj Sharma – ARC Centre of Excellence in Exciton Science, Department of Materials Science and Engineering, Monash University, Clayton Campus, Melbourne, Victoria 3800, Australia; orcid.org/0000-0001-5215-9740; Email: manoj.sharma@monash.edu

Authors

Ke Wang – College of Physics and Electronic Engineering, Chongqing Normal University, Chongqing 401331, China

Yadong Han – College of Physics and Electronic Engineering, Chongqing Normal University, Chongqing 401331, China

Zhenzhong Lian – College of Physics and Electronic Engineering, Chongqing Normal University, Chongqing 401331, China; orcid.org/0000-0003-1168-2388

Songyan Hou – Guangzhou Institute of Technology, Xidian University, Guangzhou 510555, China

Chang Cao – ARC Centre of Excellence in Exciton Science, Department of Materials Science and Engineering, Monash University, Clayton Campus, Melbourne, Victoria 3800, Australia

Hilmi Volkan Demir – LUMINOUS! Centre of Excellence for Semiconductor Lighting and Displays, School of Electrical and Electronic Engineering, Nanyang Technological University, Singapore 639798, Singapore; Department of Electrical and Electronics Engineering and Department of Physics, UNAM-Institute of Materials Science and Nanotechnology, Bilkent University, Ankara 06800, Turkey; orcid.org/0000-0003-1793-112X

Jacek J. Jasieniak – ARC Centre of Excellence in Exciton Science, Department of Materials Science and Engineering, Monash University, Clayton Campus, Melbourne, Victoria 3800, Australia; orcid.org/0000-0002-1608-6860

Complete contact information is available at: <https://pubs.acs.org/doi/10.1021/acsphotonics.5c02900>

Funding

This work was supported by the National Key Research and Development Program (2023YFB3611100), the National Natural Science Foundation of China under Grant 12304032&62205041, the Scientific and Technology Research Program of Chongqing Municipal Education Commission (KJQN202300527). M.S. and J.J. acknowledge funding through the Australian Research Council (DP260103767). H.V.D. gratefully acknowledges TÜBA – Turkish Academy of Sciences.

Notes

The authors declare no competing financial interest.

REFERENCES

- (1) Ithurria, S.; Tessier, M. D.; Mahler, B.; Lobo, R. P. S. M.; Dubertret, B.; Efros, A. Colloidal nanoplatelets with two-dimensional electronic structure. *Nat. Mater.* **2011**, *10*, 936–941.
- (2) Nasilowski, M.; Mahler, B.; Lhuillier, E.; Ithurria, S.; Dubertret, B. Two-Dimensional Colloidal Nanocrystals. *Chem. Rev.* **2016**, *116*, 10934–10982.
- (3) Lhuillier, E.; Pedetti, S.; Ithurria, S.; Nadal, B.; Heuclin, H.; Dubertret, B. Two-Dimensional Colloidal Metal Chalcogenides Semiconductors: Synthesis, Spectroscopy, and Applications. *Acc. Chem. Res.* **2015**, *48*, 22–30.
- (4) Ithurria, S.; Dubertret, B. Quasi 2D colloidal CdSe platelets with thicknesses controlled at the atomic level. *J. Am. Chem. Soc.* **2008**, *130*, 16504–16505.
- (5) Yu, J.; Dang, C. Colloidal Metal Chalcogenide Quantum Wells for Laser Applications. *Cell Rep. Phys. Sci.* **2021**, *2*, 100308.
- (6) She, C.; Fedin, I.; Dolzhenkov, D. S.; Dahlberg, P. D.; Engel, G. S.; Schaller, R. D.; Talapin, D. V. Red, Yellow, Green, and Blue Amplified Spontaneous Emission and Lasing Using Colloidal CdSe Nanoplatelets. *ACS Nano* **2015**, *9*, 9475–9485.
- (7) Hu, A.; Bai, P.; Zhu, Y.; Tang, Z.; Xiao, L.; Gao, Y. Controlled core/crown growth enables blue-emitting colloidal nanoplatelets with efficient and pure photoluminescence. *Small* **2022**, *18*, 2204120.
- (8) Bai, P.; Hu, A.; Deng, Y.; Tang, Z.; Yu, W.; Hao, Y.; Yang, S.; Zhu, Y.; Xiao, L.; Jin, Y.; Gao, Y. CdSe/CdSeS nanoplatelet light-emitting diodes with ultrapure green color and high external quantum efficiency. *J. Phys. Chem. Lett.* **2022**, *13*, 9051–9057.
- (9) Gao, H.; Luo, D.; Ren, Y.; Fang, W.; Zhou, Y.; Liao, J.; Yu, J.; Liao, S.; Ng, K. W.; Wang, S.-P.; et al. The Rise of Colloidal Quantum Well Light-Emitting Diodes. *Adv. Funct. Mater.* **2025**, *35*, 2422377.
- (10) Zhu, Y.; Deng, Y.; Bai, P.; Wu, X.; Yao, Y.; Liu, Q.; Qiu, J.; Hu, A.; Tang, Z.; Yu, W.; et al. Highly Efficient Light-Emitting Diodes Based on Self-Assembled Colloidal Quantum Wells. *Adv. Mater.* **2023**, *35* (49), 2305382.
- (11) Zhu, Y.; Lu, X.; Qiu, J.; Bai, P.; Hu, A.; Yao, Y.; Liu, Q.; Li, Y.; Yu, W.; Li, Y.; et al. High-Performance Green and Blue Light-Emitting Diodes Enabled by CdZnSe/ZnS Core/Shell Colloidal Quantum Wells. *Adv. Mater.* **2025**, *37*, 2414631.
- (12) Fang, W.; Yu, J.; Liao, S.; Gao, H.; Li, X.; Liu, B. Emergence of colloidal quantum well-based optoelectronics. *Cell Rep. Phys. Sci.* **2024**, *5*, 101936.
- (13) Hu, A.; Ma, L.; Yang, X.; Yao, Y.; Zhu, Y.; Qiu, J.; Wang, S.; Lu, C.; Gao, Y. Colloidal II–VI nanoplatelets for optoelectronic devices: Progress and perspectives. *Nano Res.* **2024**, *17*, 10412.
- (14) Lhuillier, E.; Dayen, J.-F.; Thomas, D. O.; Robin, A.; Doudin, B.; Dubertret, B. Nanoplatelets Bridging a Nanotrench: A New Architecture for Photodetectors with Increased Sensitivity. *Nano Lett.* **2015**, *15* (3), 1736.
- (15) Saitov, S. R.; Smirnov, A. M.; Saidzhonov, B. M.; Vasiliev, R. B.; Aleksandrov, A. E.; Tameev, A. R.; Snigirev, G. O.; Mantsevich, V. N. Giant Photoconductivity of Annealed CdSe/CdS Nanoplatelet Films. *ACS Photonics* **2025**, *12* (4), 2088–2096.
- (16) Dutta, A.; Medda, A.; Bera, R.; Sarkar, K.; Sain, S.; Kumar, P.; Patra, A. Hybrid Nanostructures of 2D CdSe Nanoplatelets for High-Performance Photodetector Using Charge Transfer Process. *ACS Appl. Nano Mater.* **2020**, *3* (5), 4717–4727.
- (17) Liu, B.; Sharma, M.; Yu, J.; Shendre, S.; Hettiarachchi, C.; Sharma, A.; Yeltik, A.; Wang, L.; Sun, H.; Dang, C.; et al. Light-emitting diodes with Cu-doped colloidal quantum wells: from ultrapure green, tunable dual-emission to white light. *Small* **2019**, *15*, 1901983.
- (18) Yu, J.; Sharma, M.; Delikanli, S.; Birowosuto, M. D.; Demir, H. V.; Dang, C. Mutual Energy Transfer in a Binary Colloidal Quantum Well Complex. *J. Phys. Chem. Lett.* **2019**, *10*, 5193–5199.
- (19) Smirnov, A. M.; Golinskaya, A. D.; Kotin, P. A.; Dorofeev, S. G.; Palyulin, V. V.; Mantsevich, V. N.; Dneprovskii, V. S. Photoluminescence and nonlinear transmission of Cu-doped CdSe quantum dots. *J. Lumin.* **2019**, *213*, 29–35.
- (20) Yu, J.; Hu, S.; Gao, H.; Delikanli, S.; Liu, B.; Jasieniak, J. J.; Sharma, M.; Demir, H. V. Observation of Phonon Cascades in CuDoped Colloidal Quantum Wells. *Nano Lett.* **2022**, *22*, 10224–10231.
- (21) Wu, P.; Yan, X.-P. Doped Quantum Dots for Chemo/biosensing and Bioimaging. *Chem. Soc. Rev.* **2013**, *42*, 5489–5521.
- (22) Yu, J.; Sharma, M.; Li, M.; Liu, B.; Hernández-Martínez, P. L.; Delikanli, S.; Sharma, A.; Altintas, Y.; Hettiarachchi, C.; Sum, T. C.; Demir, H. V.; Dang, C. Efficient Generation of Emissive Many-Body Correlations in Copper-Doped Colloidal Quantum Wells. *Cell Rep. Phys. Sci.* **2022**, *3*, 101049.
- (23) Bradshaw, L. R.; Knowles, K. E.; McDowall, S.; Gamelin, D. R. Nanocrystals for Luminescent Solar Concentrators. *Nano Lett.* **2015**, *15*, 1315–1323.
- (24) Volnianska, O.; Szymura, M.; Mikulski, J.; Kłopotowski, Ł. How Surface Proximity to Copper Dopants Affects Photoluminescence of CdSe Colloidal Quantum Dots. *J. Phys. Chem. C* **2021**, *125* (330), 16827–16836.
- (25) Viswanatha, R.; Brovelli, S.; Pandey, A.; Crooker, S. A.; Klimov, V. I. Copper-doped inverted core/shell nanocrystals with “permanent” optically active holes. *Nano Lett.* **2011**, *11* (11), 4753–4758.
- (26) Brovelli, S.; Galland, C.; Viswanatha, R.; Klimov, V. I. Tuning Radiative Recombination in Cu-Doped Nanocrystals via Electrochemical Control of Surface Trapping. *Nano Lett.* **2012**, *12*, 4372–4379.
- (27) Hughes, K. E.; Hartstein, K. H.; Gamelin, D. R. Photodoping and Transient Spectroscopies of Copper-Doped CdSe/CdS Nanocrystals. *ACS Nano* **2018**, *12*, 718.
- (28) Whitham, P. J.; Knowles, K. E.; Reid, P. J.; Gamelin, D. R. Photoluminescence Blinking and Reversible Electron Trapping in Copper-Doped CdSe Nanocrystals. *Nano Lett.* **2015**, *15*, 4045–4051.
- (29) Nelson, H. D.; Li, X.; Gamelin, D. R. Computational Studies of the Electronic Structures of Copper-Doped CdSe Nanocrystals: Oxidation States, Jahn–Teller Distortions, Vibronic Bandshapes, and Singlet-Triplet Splittings. *J. Phys. Chem. C* **2016**, *120* (10), 5714–5723.
- (30) Dutta, A.; Bera, R.; Ghosh, A.; Patra, A. Ultrafast Carrier Dynamics of Photo-Induced Cu-Doped CdSe Nanocrystals. *J. Phys. Chem. C* **2018**, *122*, 16992–17000.
- (31) Maiti, S.; Dana, J.; Jadhav, Y.; Debnath, T.; Haram, S. K.; Ghosh, H. N. Electrochemical Evaluation of Dopant Energetics and the Modulation of Ultrafast Carrier Dynamics in Cu-Doped CdSe Nanocrystals. *J. Phys. Chem. C* **2017**, *121* (48), 27233–27240.
- (32) Yu, J.; Demir, H. V.; Sharma, M. Optical signatures of lattice strain in chemically doped colloidal quantum wells. *Nat. Commun.* **2025**, *16*, 823.
- (33) Smirnov, A. M.; Golinskaya, A. D.; Kotin, P. A.; Dorofeev, S. G.; Zharkova, E. V.; Palyulin, V. V.; Mantsevich, V. N.; Dneprovskii, V. S. Damping of Cu-Associated Photoluminescence and Formation of Induced Absorption in Heavily Cu-Doped CdSe Quantum Dots. *J. Phys. Chem. C* **2019**, *123* (45), 27986–27992.
- (34) Yu, J.; Sharma, M.; Wang, Y.; Delikanli, S.; Baruj, H. D.; Sharma, A.; Demir, H. V.; Dang, C. Modulating Emission Properties in a Host–Guest Colloidal Quantum Well Superlattice. *Adv. Optical Mater.* **2022**, *10*, 2101756.

- (35) Jiang, T.; Song, J.; Wang, H.; Ye, X.; Wang, H.; Zhang, W.; Yang, M.; Xia, R.; Zhu, L.; Xu, X. Aqueous Synthesis of Color Tunable Cu Doped Zn-In-S/ZnS Nanoparticles in the Whole Visible Region for Cellular Imaging. *J. Mater. Chem. B* **2015**, *3*, 2402–2410.
- (36) Yu, J.; Sharma, M.; Li, M.; Delikanli, S.; Sharma, A.; Taimoor, M.; Altintas, Y.; McBride, J. R.; Kusserow, T.; Sum, T.-C.; et al. Low-Threshold Lasing from Copper-Doped CdSe Colloidal Quantum Wells. *Laser Photonics Rev.* **2021**, *15*, 2100034.
- (37) Hassan, A.; Zhang, X.; Liu, X.; Rowland, C. E.; Jawaid, A. M.; Chattopadhyay, S.; Gulec, A.; Shamirian, A.; Zuo, X.; Klie, R. F.; Schaller, R. D.; Snee, P. T. Charge carriers modulate the bonding of semiconductor nanoparticle dopants as revealed by time-resolved X-ray spectroscopy. *ACS Nano* **2017**, *11*, 10070–10076.
- (38) Jawaid, A. M.; Chattopadhyay, S.; Wink, D. J.; Page, L. E.; Snee, P. T. Cluster-Seeded Synthesis of Doped CdSe: Cu₄ Quantum Dots. *ACS Nano* **2013**, *7* (4), 3190–3197.
- (39) Sharma, M.; Gungor, K.; Yeltik, A.; Olutas, M.; Guzelturk, B.; Kelestemur, Y.; Erdem, T.; Delikanli, S.; McBride, J. R.; Demir, H. V. Near-Unity Emitting Copper-Doped Colloidal Semiconductor Quantum Wells for Luminescent Solar Concentrators. *Adv. Mater.* **2017**, *29*, 1700821.
- (40) Sharma, M.; Olutas, M.; Yeltik, A.; Kelestemur, Y.; Sharma, A.; Delikanli, S.; Guzelturk, B.; Gungor, K.; McBride, J. R.; Demir, H. V. Understanding the Journey of Dopant Copper Ions in Atomically Flat Colloidal Nanocrystals of CdSe Nanoplatelets Using Partial Cation Exchange Reactions. *Chem. Mater.* **2018**, *30*, 3265–3275.
- (41) Fuhr, A. S.; Yun, H. J.; Makarov, N. S.; Li, H.; McDaniel, H.; Klimov, V. I. Light Emission Mechanisms in CuInS₂ Quantum Dots Evaluated by Spectral Electrochemistry. *ACS Photonics* **2017**, *4*, 2425–2435.
- (42) Fuhr, A.; Yun, H. J.; Crooker, S. A.; Klimov, V. I. Spectroscopic and Magneto-Optical Signatures of Cu¹⁺ and Cu²⁺ Defects in Copper Indium Sulfide Quantum Dots. *ACS Nano* **2020**, *14*, 2212–2223.
- (43) Jara, D. H.; Stampelcoskie, K. G.; Kamat, P. V. Two Distinct Transitions in CuInS₂ Quantum Dots. Bandgap versus Sub-Bandgap Excitations in Copper-Deficient Structures. *J. Phys. Chem. Lett.* **2016**, *7*, 1452–1459.
- (44) Pinchetti, V.; Di, Q.; Lorenzon, M.; Camellini, A.; Fasoli, M.; Zavelani-Rossi, M.; Meinardi, F.; Zhang, J.; Crooker, S. A.; Brovelli, S. Excitonic pathway to photoinduced magnetism in colloidal nanocrystals with nonmagnetic dopants. *Nat. Nanotechnol.* **2018**, *13* (2), 145–151.
- (45) Zaffalon, M. L.; Pinchetti, V.; Camellini, A.; Vikulov, S.; Capitani, C.; Bai, B.; Xu, M.; Meinardi, F.; Zhang, J.; Manna, L.; et al. Intrinsic and extrinsic exciton recombination pathways in AgInS₂ colloidal nanocrystals. *Energy Mater. Adv.* **2021**, *2021*, 1959321.
- (46) van der Stam, W.; de Graaf, M.; Gudjonsdottir, S.; Geuchies, J. J.; Dijkema, J. J.; Kirkwood, N.; Evers, W. H.; Longo, A.; Houtepen, A. J. Tuning and Probing the Distribution of Cu⁺ and Cu²⁺ Trap States Responsible for Broad-Band Photoluminescence in CuInS₂ Nanocrystals. *ACS Nano* **2018**, *12*, 11244–11253.
- (47) Qin, Z.; Zhang, C.; Chen, L.; Yu, T.; Wang, X.; Xiao, M. Electrical switching of optical gain in perovskite semiconductor nanocrystals. *Nano Lett.* **2021**, *21*, 7831–7838.
- (48) Scott, R.; Achtstein, A. W.; Prudnikau, A. V.; Antanovich, A.; Siebbeles, L. D. A.; Artemyev, M.; Woggon, U. Time-Resolved Stark Spectroscopy in CdSe Nanoplatelets: Exciton Binding Energy, Polarizability, and Field-Dependent Radiative Rates. *Nano Lett.* **2016**, *16* (10), 6576–6583.
- (49) Yu, J.; Shendre, S.; Koh, W.-K.; Liu, B.; Li, M.; Hou, S.; Hettiarachchi, C.; Delikanli, S.; Hernandez-Martinez, P.; Birowosuto, M. D.; et al. Electrically Control Amplified Spontaneous Emission in Colloidal Quantum Dots. *Sci. Adv.* **2019**, *5*, No. eaav3140.
- (50) Elliott, R. J. Intensity of optical absorption by excitons. *Phys. Rev.* **1957**, *108*, 1384–1389.
- (51) Grim, J. Q.; et al. Continuous-wave biexciton lasing at room temperature using solution-processed quantum wells. *Nat. Nanotechnol.* **2014**, *9*, 891–895.
- (52) Yu, J.; Hou, S.; Sharma, M.; Tobing, L. Y. M.; Song, Z.; Delikanli, S.; Hettiarachchi, C.; Zhang, D.; Fan, W.; Birowosuto, M. D.; et al. Strong Plasmon-Wannier Mott Exciton Interaction with High Aspect Ratio Colloidal Quantum Wells. *Matter* **2020**, *2*, 1550–1563.
- (53) Yang, Y.; et al. Observation of a hot-phonon bottleneck in lead-iodide perovskites. *Nat. Photonics* **2016**, *10*, 53.
- (54) Wang, L.; Xiang, D.; Gao, K.; Wang, J.; Wu, K. Colloidal N-Doped CdSe and CdSe/ZnS Nanoplatelets. *J. Phys. Chem. Lett.* **2021**, *12*, 11259–11266.
- (55) Boehme, S. C.; Azpiroz, J. M.; Aulin, Y. V.; Grozema, F. C.; Vanmaekelbergh, D.; Siebbeles, L. D. A.; Infante, I.; Houtepen, A. J. Density of Trap States and Auger-Mediated Electron Trapping in CdTe Quantum-Dot Solids. *Nano Lett.* **2015**, *15*, 3056–3066.
- (56) Yang, L.; Knowles, K. E.; Gopalan, A.; Hughes, K. E.; James, M. C.; Gamelin, D. R. One-Pot Synthesis of Monodisperse Colloidal Copper-Doped CdSe Nanocrystals Mediated by Ligand-Copper Interactions. *Chem. Mater.* **2016**, *28*, 7375–7384.
- (57) Yu, J.; Sarma, M.; Li, M.; Hernandez-Martinez, P. L.; Delikanli, S.; Sarma, A.; Altintas, Y.; Hettiarachchi, C.; Sum, T.; Demir, H. V. et al. Sustained biexciton emission in colloidal quantum wells assisted by dopant-host interaction. *arXiv* **2019**.