

Space-confined triplet-singlet energy transfer enables high-efficiency deep-red afterglow in carbon dot hybrids

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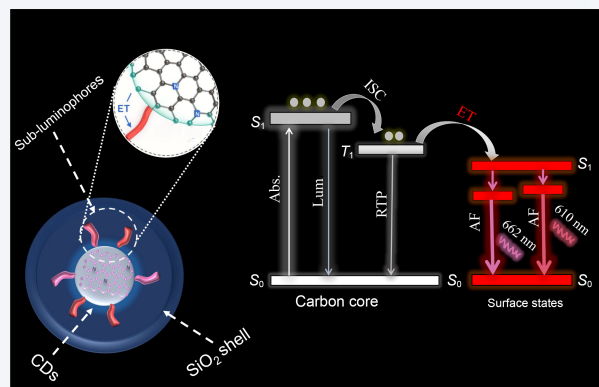
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ABSTRACT: Deep-red afterglow materials with high emission efficiency remain fundamentally limited by inefficient intersystem crossing (ISC) and significant nonradiative decay of triplet excitons under solid-state conditions. Herein, we establish a space-confined triplet-singlet energy transfer (ET) design to achieve efficient long-wavelength afterglow emission in metal-free carbon dot hybrids. Urea-induced heteroatom engineering introduces (n, π^*) states that facilitate ISC and increase triplet population, while an (3-aminopropyl) triethoxysilane-derived siloxane network rigidifies the microenvironment and suppresses vibrational relaxation, thereby stabilizing triplet excitons. Meanwhile, surface-state modulation enables favorable triplet energy alignment between the carbon core and surface-associated emissive centers, facilitating efficient triplet-mediated ET. This cooperative regulation results in bright deep-red afterglow centered at 662 nm with a photoluminescence quantum yield of 45.2%. Comparative investigations with red-emissive counterparts reveal that surface-state modulation and molecular rigidification play complementary roles in wavelength tunability and emission efficiency. The resulting materials demonstrate potential in time-resolved optical encryption and persistent afterglow lighting. This work provides mechanistic insight into triplet regulation in confined carbon systems and suggests a viable strategy for improving long-wavelength metal-free afterglow performance.



KEYWORDS: deep-red afterglow, carbon dots, triplet-singlet energy transfer, surface-state modulation, afterglow lighting

1 Introduction

Long-wavelength afterglow materials with high luminescence efficiency have attracted increasing attention due to their potential applications in artificial lighting, information encryption, bioimaging, and long-wavelength display technologies [1–4]. In particular, red luminophores serve as critical down-conversion

components in white light-emitting devices, improving color-rendering performance and spectral balance [5]. Long-wavelength emissive materials are especially desirable for biological imaging and detection because photons penetrate deeply into biological tissues with minimal scattering and autofluorescence interference, enabling high signal-to-noise detection [4]. Despite these advantages, current long-wavelength emitters largely rely on heavy-metal-based quantum dots, perovskites, or small-molecule organic phosphors [6–8]. The toxicity of heavy metals and the limited environmental stability of perovskites have motivated the development of metal-free and sustainable persistent luminescence systems capable of operating efficiently under ambient conditions.

Organic afterglow materials exploit long-lived triplet excitons to

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achieve persistent emission through strategies such as molecular rigidification, host-guest assembly, and crystallization engineering [9–11]. However, extending afterglow emission into the red and long-wavelength spectral regions remains fundamentally challenging. Two intrinsic limitations dominate this regime, inefficient intersystem crossing (ISC), which restricts triplet population, and accelerated nonradiative decay of low-energy triplet excitons governed by the energy gap law [6–8, 12, 13]. Although spatial confinement within rigid matrices has been shown to effectively stabilize triplet excitons and enable efficient blue-to-green afterglow emission [2, 14], achieving both high efficiency and long-wavelength emission in a metal-free system remains difficult because triplet generation and stabilization are rarely regulated simultaneously.

Carbon dots (CDs) provide a promising platform to address these challenges [15–19]. Their sp^2 -hybridized carbon cores, combined with tunable surface functional groups, offer opportunities to modulate spin-orbit coupling (SOC) and excited-state dynamics through heteroatom engineering [20–22]. Moreover, CDs inherently contain multiple surface luminescent states with distinct energy levels, enabling engineered energy transfer (ET) among emissive centers. Rather than relying solely on direct triplet emission, rational regulation of triplet migration pathways, particularly triplet-to-singlet ET between the carbon core and surface-associated emissive centers, may enable decoupled control over triplet harvesting and emission wavelength. Such a mechanism could provide an effective route toward efficient long-wavelength afterglow in carbon-based systems (Fig. 1) [23, 24].

Herein, inspired by this concept, we established a space-confined triplet-singlet ET strategy in CD hybrids. Seed CDs are synthesized via hydrothermal treatment of Nile red or Rhodamine B (RhB) with (3-aminopropyl)triethoxysilane (APTES), followed by urea-assisted condensation to construct a rigid siloxane confinement network. Urea-induced heteroatom incorporation introduces (n, π^*) states that are expected to facilitate ISC, while the cross-linked siloxane framework suppresses vibrational relaxation and stabilizes triplet excitons. The resulting confined triplet reservoir is proposed to enable energy migration from the carbon core to surface-associated emissive groups, producing deep-red (DR) afterglow and red (R) afterglow emission centered at 662 and 610 nm with steady-state photoluminescence quantum yields (PLQYs) of 45.28% and 98.64%, respectively. This dual-regulation approach integrates triplet population enhancement and nonradiative suppression within a single architecture, providing a viable pathway for designing high-efficiency long-wavelength metal-free afterglow materials.

2 Experimental section

2.1 Materials

Urea (99.5%), APTES (98%), ethanol (99.7%), RhB (95%), and Nile red (95%) were purchased from Aladdin Ltd. (Shanghai, China). All the reagents were used without further purification.

2.2 Preparation of DR-CDs

The seed DR-CDs were synthesized by dissolving 1 mL of APTES and 2 mg of Nile red in 10 mL of ethanol and the solution was transferred into a 50 mL polytetrafluoroethylene-lined stainless-steel autoclave and heated at 160 °C for 6 h (Fig. S1 in the Electronic Supplementary Material (ESM)). The obtained solution was cooled down to room temperature and centrifuged at 8000 $r \cdot min^{-1}$ for 10 min and the liquid supernatant was collected to obtain DR-seed CDs.

For the hybridization process, 2 mL of DR-seed CD solution and 500 mg of urea were mixed in 3 mL of deionized water in a vial, which was then placed in an oil bath at 150 °C for 2 h to obtain DR-CD powder (Fig. S2 in the ESM), which was used for further characterization and applications.

2.3 Preparation of R-CDs

The synthesis of R-CDs followed the same protocol as for DR-CDs, but with RhB and ethanol used in place of Nile-red as the organic precursor and deionized water substituting for the ethanol solvent.

2.4 Equipment and characterization

The morphology and lattice structures of the synthesized materials were characterized using transmission electron microscopy (TEM) and high-resolution TEM (HRTEM) on a JEOL JEM-2100F microscope. Powder X-ray diffraction (XRD) patterns were collected on a Rigaku Ultima IV diffractometer with $Cu K\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$) to evaluate the crystallographic features of the samples. Fourier transform infrared (FT-IR) spectra were recorded on a Nicolet 6700 spectrometer to identify surface functional groups and bonding characteristics. The surface elemental composition and chemical states were analyzed by X-ray photoelectron spectroscopy (XPS) using a VG Scientific ESCALAB 220i-XL spectrometer equipped with $Al K\alpha$ radiation. Ultraviolet-visible (UV-vis) absorption spectra were measured with a Shimadzu UV-3101PC spectrophotometer. Steady-state photoluminescence (PL) and excitation spectra were obtained using a Hitachi F-7000 spectrofluorometer. Afterglow emission spectra were recorded with a QE Pro spectrometer (Ocean Insight) with a delay time of 1 ms after termination of the excitation source. The

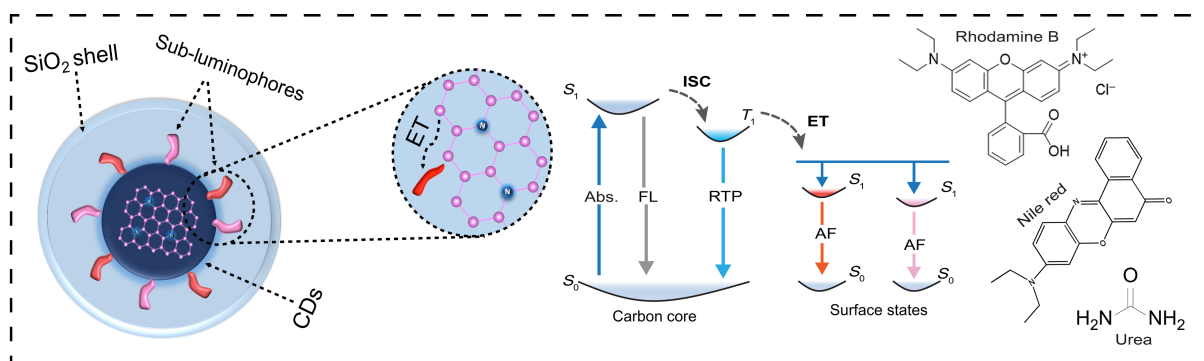


Figure 1 Schematic illustration of the design principle and proposed triplet-mediated energy-transfer process in a space-confined CD-based hybrid system.

PLQYs and afterglow lifetimes were measured using an FLS5 fluorescence spectrometer (Edinburgh Instruments). All optical measurements were performed at room temperature under ambient atmosphere unless otherwise specified.

3 Results and discussion

3.1 Material design

Although embedding CDs into rigid matrices can partially suppress vibrational relaxation, most reported CD-based systems still exhibit negligible long-lived afterglow under ambient conditions. This limitation is largely attributed to insufficient triplet population arising from limited ISC efficiency [22–25]. Therefore, efficient long-wavelength afterglow requires the simultaneous enhancement of triplet generation and suppression of nonradiative triplet dissipation.

To increase triplet population, urea was introduced as a multifunctional precursor to induce heteroatom incorporation and surface passivation. The formation of C=O, C=N, and $-NH_2$ functional groups generates (n, π^*) states that are expected to enhance SOC and facilitate ISC from singlet to triplet states. This heteroatom-mediated regulation is thus expected to increase triplet yield within the carbon core [26]. Consistently, control samples prepared without urea exhibit only negligible afterglow under ambient conditions, together with markedly shortened delayed lifetimes and significantly reduced quantum yields (Fig. S8 in the ESM), highlighting the essential role of the second-step urea-assisted condensation. Subsequently, APTES was employed to construct a cross-linked siloxane network through a process of condensation and hydrolysis. The resulting three-dimensional framework strengthens the microenvironment around the CDs, effectively suppressing intramolecular rotations and vibrational relaxation, two dominant pathways for triplet deactivation in organic systems. This structural confinement helps stabilize the triplet excitons, extending their lifetime.

To enable wavelength tuning, RhB and Nile red were incorporated as surface-associated emissive centers. Through rational triplet energy alignment between the carbon core and these sub-luminophores, triplet-mediated ET is proposed to occur. This architecture establishes a confined triplet reservoir capable of directing energy migration toward long-wavelength emissive states.

3.2 Photophysical properties

Under 365 nm excitation, both R-CDs and DR-CDs powders exhibit intense visible emission. After the 365 nm UV lamp is turned off, bright and persistent afterglow emission is clearly observed, as shown in Fig. 2(a), and Videos S1 and S2 in the ESM. Notably, the afterglow of R-CDs remains visible even under daylight conditions (Fig. S3 in the ESM), indicating strong emission intensity and environmental robustness. As shown in Figs. 2(b) and 2(c), under excitation, the steady-state PL spectra of R-CDs and DR-CDs exhibit dual emission bands centered at approximately 440/610 and 440/662 nm, respectively. After the excitation is removed, the short-wavelength emission slightly red-shifts to 452 nm, whereas the long-wavelength emission bands remain essentially unchanged and overlap with those observed in the steady-state spectra. This spectral invariance suggests that the long-wavelength afterglow originates from stabilized excited states rather than transient fluorescence.

The chromaticity evolution further corroborates the spectral behavior. The Commission Internationale de l'Eclairage (CIE) chromaticity coordinates of the steady-state PL are (0.62, 0.34) for R-CDs and (0.44, 0.27) for DR-CDs. Upon removal of excitation, the corresponding afterglow coordinates shift slightly to (0.63, 0.34) and (0.45, 0.23), respectively (Fig. S6 in the ESM). This subtle variation is consistent with the red shift of the short-wavelength component and reflects the increased contribution of triplet-mediated emission to the overall afterglow output.

To further probe the afterglow emission mechanism, time-resolved decay profiles were collected and fitted using a multi-exponential function (Figs. 2(d) and 2(e)) according to Eq. (1) [27]

$$\tau_{\text{avg}} = \sum \alpha_i \tau_i^2 / \sum \alpha_i \tau_i \quad (1)$$

The resulting average lifetimes are 354 (452 nm) and 178 ms (610 nm) for R-CDs, and 270 (452 nm) and 240 ms (662 nm) for DR-CDs. These millisecond-scale lifetimes are consistent with the involvement of long-lived triplet-mediated processes in the afterglow emission. The extracted lifetime components and average lifetimes at the emission maxima are summarized in Table S1 in the ESM.

Spectroscopic analysis further supports a triplet-mediated energy-transfer mechanism. The spectral overlap between the short-wavelength phosphorescence component and the absorption spectra of seed CDs (Fig. S9 in the ESM) suggests the possibility of triplet energy migration [28, 29]. Afterglow excitation spectra monitored at 610 (R-CDs) and 662 nm (DR-CDs) show strong overlap between the 400 and 550 nm region and the short-wavelength phosphorescence band, particularly for R-CDs (Fig. 2(f)). These observations are consistent with a space-confined triplet-singlet ET pathway.

The long-wavelength afterglow is more consistent with a space-confined triplet-mediated energy-transfer process than with conventional room-temperature phosphorescence (RTP) or thermally activated delayed fluorescence (TADF). In RTP, delayed emission originates directly from triplet states, whereas TADF relies on reverse intersystem crossing. In the present system, the delayed emission is attributed to energy transfer from confined triplet states in the carbon core to surface-associated emissive states. The nearly unchanged long-wavelength emission at 77 K (Fig. S7 in the ESM), together with the spectral overlap between carbon-core phosphorescence and surface-state absorption (Fig. S9 in the ESM), is consistent with this interpretation but is not used as a sole criterion.

The steady-state PLQYs reach 98.6% for R-CDs and 45.2% for DR-CDs under 365 nm excitation (Figs. S4 and S5 in the ESM). It should be noted that the reported PLQYs correspond to steady-state PL under continuous excitation, whereas the afterglow performance is evaluated through delayed-emission spectra and lifetime measurements. Compared with representative long-wavelength afterglow systems reported previously, which often suffer from limited emission wavelength or reduced efficiency due to increased nonradiative losses in the long-wavelength region, the present materials exhibit a more balanced performance (Fig. 2(g)). In particular, the system combines (i) long-wavelength delayed emission (~ 662 nm), (ii) relatively high PLQYs, and (iii) stable emission under continuous irradiation together with good thermal robustness, indicating an effective balance among emission wavelength, efficiency, and stability.

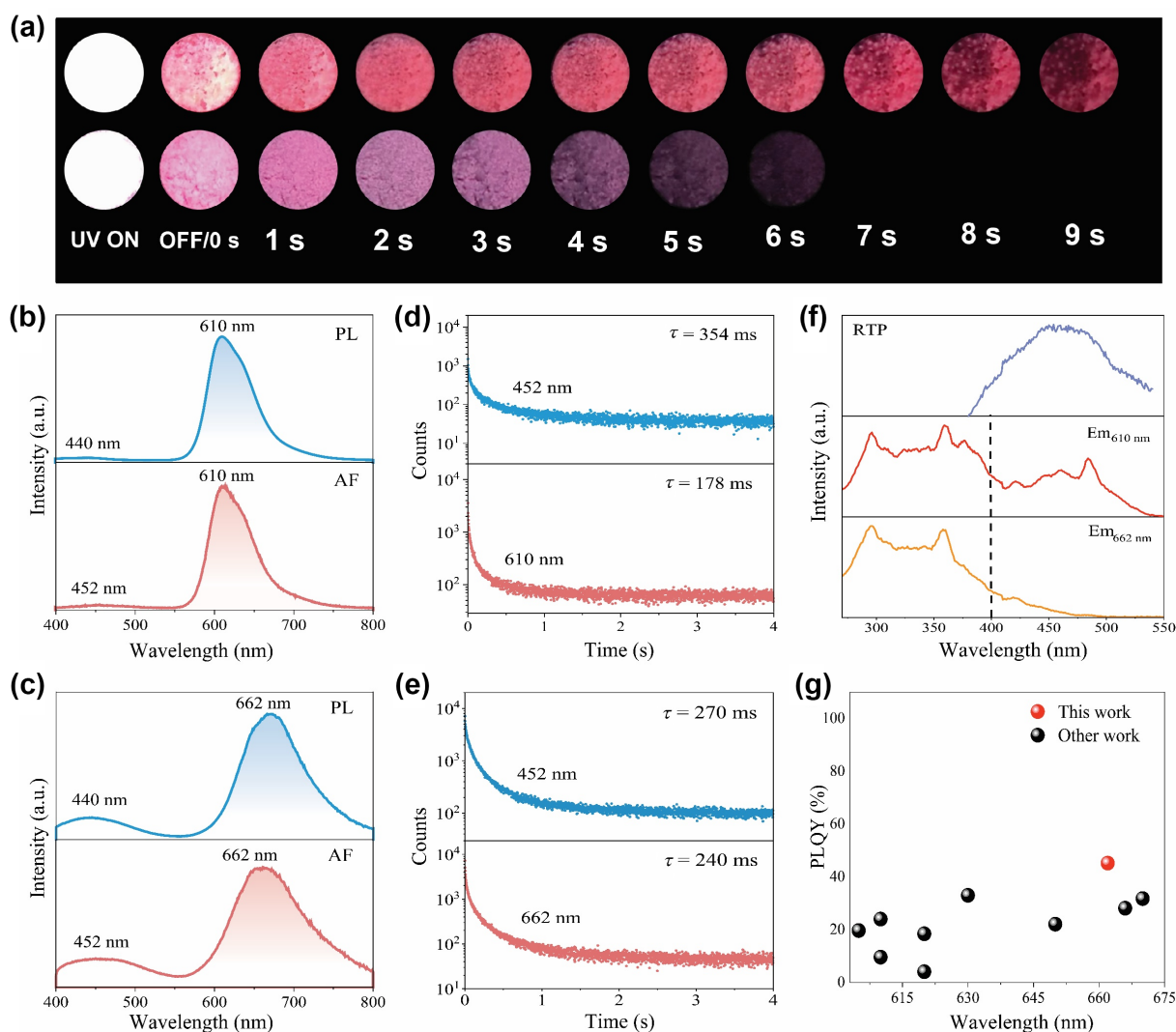


Figure 2 Photophysical properties of R-CDs and DR-CDs. (a) Photographs of the powders under 365 nm excitation (UV on) and after removal of the excitation source (0–9 s). ((b) and (c)) PL and afterglow emission spectra of (b) R-CDs and (c) DR-CDs under 365 nm excitation. ((d) and (e)) Afterglow decay profiles of (d) R-CDs and (e) DR-CDs monitored at representative wavelengths, with the extracted average lifetimes indicated. (f) Spectral comparison showing the short-wavelength phosphorescence-like component (top) and afterglow excitation spectra monitored at $E_m = 610$ nm (R-CDs, middle) and $E_m = 662$ nm (DR-CDs, bottom). (g) Comparison of PLQYs of reported long-wavelength afterglow materials (605–675 nm) and the present work.

3.3 Morphology and chemical structure

To correlate structural features with photophysical performance, detailed morphological and chemical analyses were conducted (Fig. 3). TEM images reveal uniform particle sizes of approximately 4 nm for R-CDs (Figs. 3(a)–3(c)) and 4 nm for DR-CDs (Figs. 3(d)–3(f)). High-resolution TEM images display well-resolved lattice fringes with an interplanar spacing of 0.21 nm, corresponding to the (100) plane of graphitic carbon [30, 31], suggesting the formation of partially crystalline carbon cores (Figs. 3(b) and 3(e)). XRD patterns (Fig. 3(g)) exhibit broad diffraction features centered around 21.6° , characteristic of amorphous SiO_2 tetrahedral networks [32]. The absence of distinct crystalline reflections suggests that the CDs are well dispersed within the siloxane matrix, which may help suppress aggregation-induced quenching and stabilize emissive centers through spatial confinement.

FT-IR spectra (Fig. 3(h)) display characteristic absorption bands associated with C=O (1693 cm^{-1}), C=N (1614 cm^{-1}), and $-\text{NH}_2$

(1570 cm^{-1}) groups [33, 34], indicating heteroatom incorporation. Broad absorption between $2800\text{--}3700\text{ cm}^{-1}$ corresponds to $-\text{CH}_2$, $-\text{NH}_2$, and $-\text{OH}$ stretching vibrations [29, 35, 36]. In addition, distinct peaks at 1136 and 1023 cm^{-1} are assigned to Si–O–Si and Si–O–C stretching modes, suggesting covalent coupling between CDs and the silica framework [33, 37].

XPS survey spectra (Fig. 3(i)) show characteristic peaks of Si 2p (103 eV), C 1s (285 eV), N 1s (399 eV), and O 1s (531 eV) [23, 38], confirming the hybrid composition. High-resolution C 1s spectra (Fig. 3(j)) reveal contributions from C=O (289 eV), C–N ($285.5\text{--}285.7\text{ eV}$), and C–Si (284.2 eV) bonds [6, 39–41]. The O 1s spectra (Fig. 3(k)) can be deconvoluted into components corresponding to C=O (531.0 eV), C–OH (531.7 eV), and Si–O ($532.2\text{--}532.4\text{ eV}$) [33, 37]. Meanwhile, Si 2p spectra (Fig. 3(l)) show peaks attributed to O–Si and C–Si bonding states [35, 42], supporting the formation of a cross-linked CD– SiO_2 hybrid network.

Collectively, these structural results are consistent with the proposed dual-regulation mechanism. Heteroatom

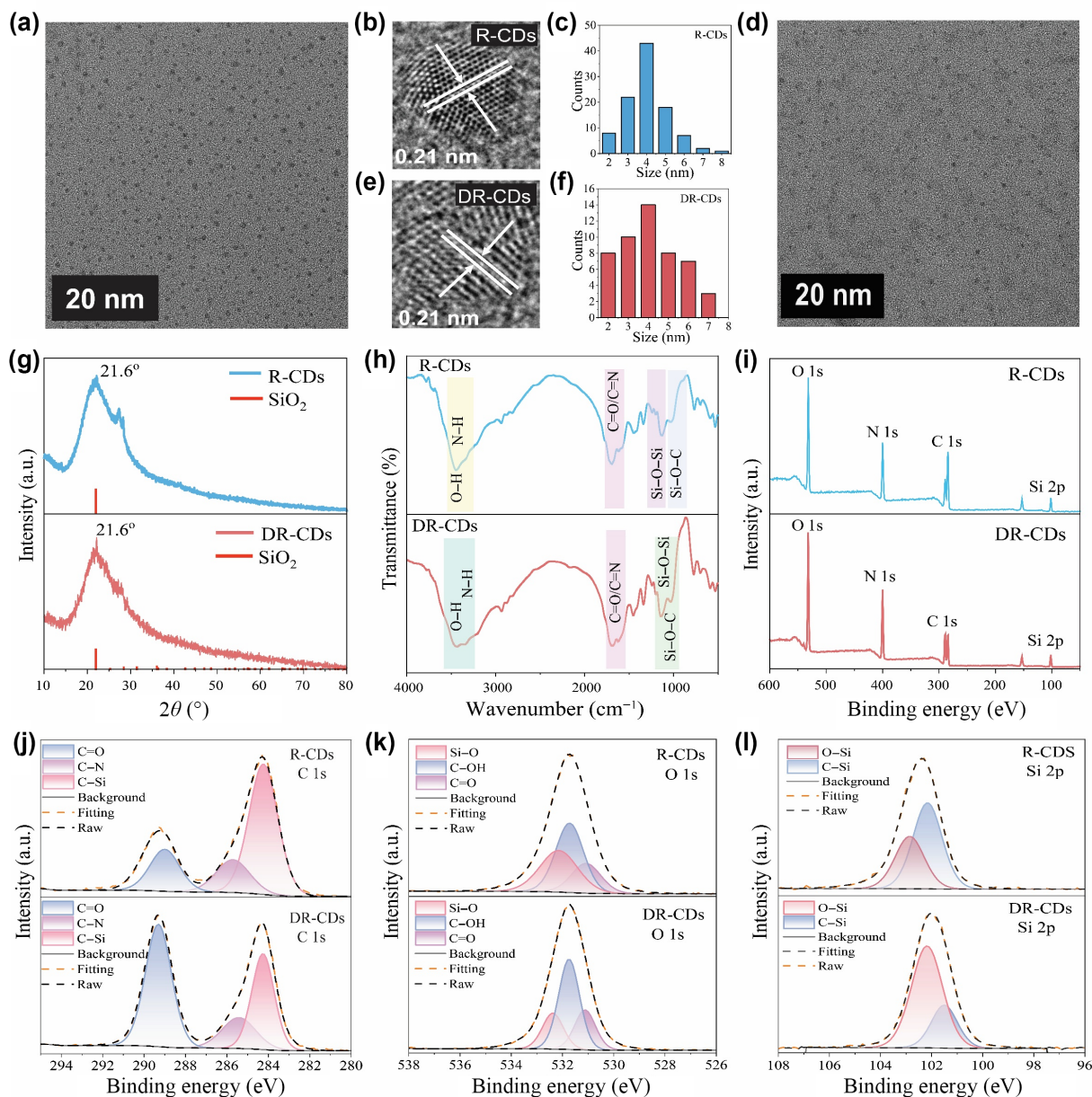


Figure 3 Structural and chemical characterization of R-CDs and DR-CDs. (a) TEM image, (b) HR-TEM image, and (c) corresponding particle size distribution of R-CDs. (d) TEM image, (e) HR-TEM image, and (f) corresponding particle size distribution of DR-CDs. (g) Powder XRD patterns of R-CDs and DR-CDs. (h) FT-IR spectra of R-CDs and DR-CDs. (i) XPS survey spectra of R-CDs and DR-CDs. (j)–(l) High-resolution XPS spectra with peak deconvolution of (j) C 1s, (k) O 1s, and (l) Si 2p.

functionalization introduces (n , π^*) states that are expected to facilitate ISC and increase triplet population [20–22], while the covalently cross-linked siloxane network imposes spatial confinement that suppresses vibrational relaxation and helps stabilize triplet excitons. The strong interfacial coupling between the carbon core and silica matrix contributes to structural rigidity, thereby supporting long-lived excited states required for persistent afterglow emission.

3.4 Mechanism

In this work, the “carbon core” refers to the graphitic CD framework that acts as the primary triplet-generating domain, whereas the “surface-associated emissive states” refer to the RhB- or

Nile red-derived chromophoric states coupled to the CD surface and responsible for the final long-wavelength delayed emission. Unlike conventional delayed-emission pathways, where the delayed signal typically arises from direct phosphorescence or reverse intersystem crossing-assisted delayed fluorescence, the present system is more appropriately described as a space-confined triplet-mediated energy-transfer process, in which the delayed emission is attributed to energy transfer to surface emissive states rather than direct radiative decay from triplet states. Upon photoexcitation, singlet excitons are initially generated within the carbon core. In the first step, APTES undergoes hydrolysis and condensation to form a cross-linked siloxane network around the CDs. This rigid microenvironment spatially confines the emissive centers, suppresses vibrational relaxation and other nonradiative decay

pathways, and thereby helps stabilize triplet excitons once they are formed [2, 6, 14]. Such confinement is essential for maintaining long-lived excited states in the hybrid system but alone is insufficient to generate efficient afterglow emission.

In the second step, urea-assisted condensation introduces heteroatom-containing surface functionalities, including C=O, C=N, and -NH_2 groups, as supported by the FT-IR and XPS results. These groups are expected to introduce accessible (n , π^*) states and are consistent with facilitating intersystem crossing, thereby contributing to triplet formation [20–22]. Consistently, control experiments (Fig. S8 in the ESM) show that samples prepared without urea exhibit comparable steady-state PL but only negligible afterglow emission under ambient conditions, together with significantly shortened delayed lifetimes and reduced quantum yields. These observations highlight the essential role of the urea-assisted second step in enabling efficient afterglow, rather than merely modifying the confined structure.

The resulting confined triplet reservoir is proposed to transfer energy to the surface-associated emissive states through favorable energy-level matching and spectral overlap, thereby giving rise to the final long-wavelength delayed emission [24, 30, 31, 37]. This assignment is supported by the spectral overlap between the short-wavelength phosphorescence component and the absorption features of the seed CDs, together with the excitation-dependent afterglow spectra.

Overall, the afterglow behavior can be understood as arising from the synergistic effect of triplet stabilization via APTES-derived spatial confinement and triplet generation facilitated with urea-induced heteroatom incorporation. This decoupled architecture spatially separates triplet generation, triplet stabilization, and emission-wavelength regulation, thereby enabling long-wavelength afterglow. Such synergistic regulation of spatial confinement and heteroatom incorporation may provide a useful strategy for designing high-performance long-wavelength metal-free afterglow materials [6–8].

3.5 Demonstration of light-emitting diode (LED) integration and information encryption applications

Both CD systems exhibit stable photoluminescence behavior under continuous 365 nm irradiation and good thermal robustness. The data shown in Fig. S10 in the ESM correspond to the stability of steady-state PL under continuous 365 nm excitation. The emission intensity was monitored at the characteristic emission peaks (610 nm for R-CDs and 662 nm for DR-CDs) as a function of irradiation time. No delay window was applied in this measurement, indicating that the recorded signal corresponds to prompt PL rather than delayed afterglow emission. The photostability was evaluated by tracking the normalized PL intensity over time under continuous irradiation. Moreover, upon heating from 298 to 368 K, the PL intensities decreased by only 14% for R-CDs and 21% for DR-CDs (Fig. S11 in the ESM), indicating good thermal robustness. Such stability is beneficial for device integration and supports the potential of these materials in long-wavelength optoelectronic applications.

To evaluate their potential in solid-state lighting, R-CDs and DR-CDs were integrated into 365 nm LED chips (Fig. 4(a)). The CD powders were uniformly coated onto the chips (Fig. S12 in the ESM), forming afterglow devices. Under UV excitation, both devices exhibit broad electroluminescence across the visible region (Figs. 4(b) and 4(c)). The R-LED achieves a luminance of

2781 $\text{cd}\cdot\text{m}^{-2}$ at 3.3 V, with an external quantum efficiency (EQE) of 1.01%, a current efficiency of $1.59\text{ cd}\cdot\text{A}^{-1}$, and a power efficiency of $1.48\text{ lm}\cdot\text{W}^{-1}$. The DR-LED achieves a luminance of $13890\text{ cd}\cdot\text{m}^{-2}$ at 3.32 V and 48 mA, with an EQE of 1.24%, a current efficiency of $0.83\text{ cd}\cdot\text{A}^{-1}$, and a power efficiency of $0.78\text{ lm}\cdot\text{W}^{-1}$. Notably, after the excitation source is turned off, both devices continue to emit visible afterglow for several seconds (Figs. 4(d) and 4(e)), suggesting efficient triplet utilization at the device level.

The operational durability of the DR-LED was further evaluated by monitoring normalized luminance decay over time (Fig. 4(f)). The T_{95} value, defined as the time required for luminance to decrease to 95% of its initial intensity ($L_0 = 1000\text{ cd}\cdot\text{m}^{-2}$), was measured to be 3.46 h, demonstrating stable device performance under continuous operation.

Beyond lighting, the excitation-dependent long-wavelength afterglow also enables time-gated optical encryption. Multicolor “ZZU” patterns prepared using R-CD and DR-CD powders show distinct fluorescence under UV illumination and persistent delayed emission after the excitation is removed (Fig. 4(g)). The spectral difference between the red and deep-red afterglow introduces a temporal dimension to information encoding, highlighting the potential of these materials for anti-counterfeiting, secure labeling, and dynamic optical storage.

4 Conclusions

In summary, we report a two-step hybridization strategy to construct CD-based siloxane composites that exhibit efficient red and deep-red afterglow through a space-confined triplet-singlet ET pathway. Urea-induced heteroatom functionalization introduces accessible (n , π^*) states that are expected to facilitate intersystem crossing, while the APTES-derived siloxane network rigidifies the local microenvironment and suppresses vibrational dissipation of triplet excitons. This dual regulation effectively helps stabilize long-lived excited states and enables energy migration from the carbon core to surface-associated emissive centers. As a result, the R-CDs and DR-CDs exhibit high steady-state PLQYs of 98.6% and 45.2%, respectively, together with bright and persistent long-wavelength afterglow. These results provide insight into the cooperative roles of heteroatom engineering and structural confinement in regulating triplet dynamics within metal-free hybrid systems. The demonstrated integration with UV LED chips and time-gated optical encryption further highlights the application potential of this design strategy for long-wavelength persistent luminescence materials.

Electronic Supplementary Material: Supplementary material (schematic illustrations of the synthesis and hybridization process, photographs and photoluminescence/afterglow spectra, quantum yield and photostability measurements, temperature-dependent spectra, LED device photographs, and tables) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94908849>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

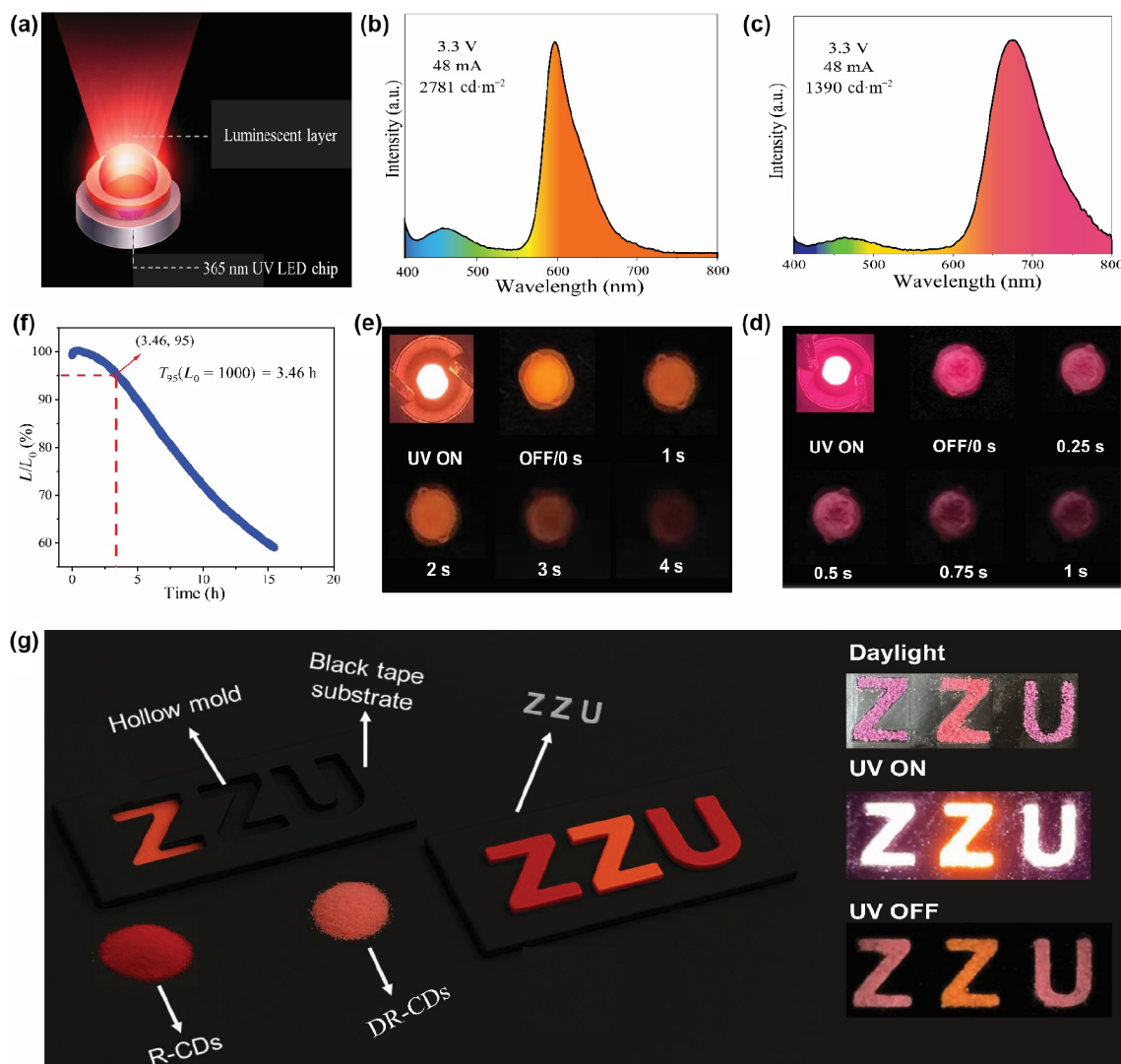


Figure 4 Device integration and optical encryption applications. (a) Schematic illustration of the CD-coated UV LED architecture. ((b) and (c) Luminescence spectra of (b) R-LED and (c) DR-LED. ((d) and (e) Photographs of (d) DR-LED and (e) R-LED under UV illumination (UV on) and after removal of the excitation source (UV off), showing the afterglow emission. (f) Operational stability of the DR-LED, showing normalized luminance (L/L_0) as a function of time, with $T_{95}(L_0 = 1000 \text{ cd}\cdot\text{m}^{-2})$ defined as the time required for the luminance to decrease to 95% of its initial value. (g) Multicolor “ZZU” patterns fabricated using R-CDs and DR-CDs for time-gated optical encryption under daylight, UV illumination, and afterglow conditions.

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Author contribution statement

U. H.: Data curation, writing manuscript. J.-Y. Z. and Q. L.: Project administration, validation, experimental design. G.-S. Z., Z.-C. Z., A. R. H., C. L., X.-F. X., and J.-H. Z.: Data curation, experimental design, methodology, writing, review and editing. C.-X. S.: Project administration, funding acquisition, validation. All the authors have approved the final manuscript.

Use of AI statement

None.

References

- [1] Xu, S.; Wang, W.; Li, H.; Zhang, J. Y.; Chen, R. F.; Wang, S.; Zheng, C.; Xing, G. C.; Song, C. Y.; Huang, W. Design of highly efficient deep-blue organic afterglow through guest sensitization and matrices rigidification. *Nat. Commun.* **2020**, *11*, 4802.
- [2] Ye, W. P.; Ma, H. L.; Shi, H. F.; Wang, H.; Lv, A. Q.; Bian, L. F.; Zhang, M.; Ma, C. Q.; Ling, K.; Gu, M. X. et al. Confining isolated chromophores for highly efficient blue phosphorescence. *Nat. Mater.* **2021**, *20*, 1539–1544.
- [3] Tan, J.; Li, Q. J.; Meng, S.; Li, Y. C.; Yang, J.; Ye, Y. X.; Tang, Z. K.; Qu, S. N.; Ren, X. D. Time-dependent phosphorescence colors from carbon dots for advanced dynamic information encryption. *Adv. Mater.* **2021**, *33*, 2006781.
- [4] Xiao, F. M.; Gao, H. Q.; Lei, Y. X.; Dai, W. B.; Liu, M. C.; Zheng, X. Y.; Cai, Z. X.; Huang, X. B.; Wu, H. Y.; Ding, D. Guest–host doped strategy for constructing ultralong-lifetime near-infrared organic phosphorescence materials for bioimaging. *Nat. Commun.* **2022**, *13*, 186.

- [5] Chen, Q. G.; Qu, L. J.; Hou, H.; Huang, J. Y.; Li, C.; Zhu, Y.; Wang, Y. K.; Chen, X. H.; Zhou, Q.; Yang, Y. et al. Long lifetimes white afterglow in slightly crosslinked polymer systems. *Nat. Commun.* **2024**, *15*, 2947.
- [6] Song, Z. J.; Shang, Y.; Lou, Q.; Zhu, J. Y.; Hu, J. H.; Xu, W.; Li, C. C.; Chen, X.; Liu, K. K.; Shan, C. X. et al. A molecular engineering strategy for achieving blue phosphorescent carbon dots with outstanding efficiency above 50%. *Adv. Mater.* **2023**, *35*, 2207970.
- [7] Jiang, K.; Wang, Y. H.; Gao, X. L.; Cai, C. Z.; Lin, H. W. Facile, quick, and gram-scale synthesis of ultralong-lifetime room-temperature-phosphorescent carbon dots by microwave irradiation. *Angew. Chem., Int. Ed.* **2018**, *57*, 6216–6220.
- [8] Shi, W. Q.; Zeng, L. L.; He, R. L.; Han, X. S.; Guan, Z. J.; Zhou, M.; Wang, Q. M. Near-unity NIR phosphorescent quantum yield from a room-temperature solvated metal nanocluster. *Science* **2024**, *383*, 326–330.
- [9] Gan, N.; Zou, X.; Qian, Z.; Lv, A. Q.; Wang, L.; Ma, H. L.; Qian, H. J.; Gu, L.; An, Z. F.; Huang, W. Stretchable phosphorescent polymers by multiphase engineering. *Nat. Commun.* **2024**, *15*, 4113.
- [10] Ma, X. K.; Zhang, W.; Liu, Z. X.; Zhang, H. Y.; Zhang, B.; Liu, Y. Supramolecular pins with ultralong efficient phosphorescence. *Adv. Mater.* **2021**, *33*, 2007476.
- [11] Kabe, R.; Adachi, C. Organic long persistent luminescence. *Nature* **2017**, *550*, 384–387.
- [12] Song, S. Y.; Liu, K. K.; Mao, X.; Cao, Q.; Li, N.; Zhao, W. B.; Wang, Y.; Liang, Y. C.; Zang, J. H.; Li, X. et al. Colorful triplet excitons in carbon nanodots for time delay lighting. *Adv. Mater.* **2023**, *35*, 2212286.
- [13] Pan, Y. T.; Li, J. Y.; Wang, X. P.; Sun, Y.; Li, J. B.; Wang, B. B.; Zhang, K. K. Highly efficient TADF-type organic afterglow of long emission wavelengths. *Adv. Funct. Mater.* **2022**, *32*, 2110207.
- [14] Zhang, X.; Zeng, M. J.; Zhang, Y. W.; Zhang, C. Y.; Gao, Z. S.; He, F.; Xue, X. D.; Li, H. H.; Li, P.; Xie, G. Z. et al. Multicolor hyperafterglow from isolated fluorescence chromophores. *Nat. Commun.* **2023**, *14*, 475.
- [15] Wang, Z. F.; Yuan, F. L.; Li, X. H.; Li, Y. C.; Zhong, H. Z.; Fan, L. Z.; Yang, S. H. 53% efficient red emissive carbon quantum dots for high color rendering and stable warm white-light-emitting diodes. *Adv. Mater.* **2017**, *29*, 1702910.
- [16] Liu, J. C.; Wang, N.; Yu, Y.; Yan, Y.; Zhang, H. Y.; Li, J. Y.; Yu, J. H. Carbon dots in zeolites: A new class of thermally activated delayed fluorescence materials with ultralong lifetimes. *Sci. Adv.* **2017**, *3*, e1603171.
- [17] Lu, S. Y.; Xiao, G. J.; Sui, L. Z.; Feng, T. L.; Yong, X.; Zhu, S. J.; Li, B. J.; Liu, Z. Y.; Zou, B.; Jin, M. X. et al. Piezochromic carbon dots with two-photon fluorescence. *Angew. Chem., Int. Ed.* **2017**, *56*, 6187–6191.
- [18] Yang, H. Y.; Liu, Y. L.; Guo, Z. Y.; Lei, B. F.; Zhuang, J. L.; Zhang, X. J.; Liu, Z. M.; Hu, C. F. Hydrophobic carbon dots with blue dispersed emission and red aggregation-induced emission. *Nat. Commun.* **2019**, *10*, 1789.
- [19] Wang, L.; Li, W. T.; Yin, L. Q.; Liu, Y. J.; Guo, H. Z.; Lai, J. W.; Han, Y.; Li, G.; Li, M.; Zhang, J. H. et al. Full-color fluorescent carbon quantum dots. *Sci. Adv.* **2020**, *6*, eabb6772.
- [20] Sun, Y. Q.; Liu, S. T.; Sun, L. Y.; Wu, S. S.; Hu, G. Q.; Pang, X. L.; Smith, A. T.; Hu, C. F.; Zeng, S. S.; Wang, W. X. et al. Ultralong lifetime and efficient room-temperature phosphorescent carbon dots through multi-confinement structure design. *Nat. Commun.* **2020**, *11*, 5591.
- [21] Li, Q. J.; Zhao, H. J.; Yang, D. Q.; Meng, S.; Gu, H. L.; Xiao, C.; Li, Y.; Cheng, D. K.; Qu, S. N.; Zeng, H. B. et al. Direct *in situ* fabrication of multicolor afterglow carbon dot patterns with transparent and traceless features via laser direct writing. *Nano Lett.* **2024**, *24*, 3028–3035.
- [22] Li, W.; Zhou, W.; Zhou, Z. S.; Zhang, H. R.; Zhang, X. J.; Zhuang, J. L.; Liu, Y. L.; Lei, B. F.; Hu, C. F. A universal strategy for activating the multicolor room-temperature afterglow of carbon dots in a boric acid matrix. *Angew. Chem., Int. Ed.* **2019**, *58*, 7278–7283.
- [23] Liang, Y. C.; Cao, Q.; Liu, K. K.; Peng, X. Y.; Sui, L. Z.; Wang, S. P.; Song, S. Y.; Wu, X. Y.; Zhao, W. B.; Deng, Y. et al. Phosphorescent carbon-nanodots-assisted Förster resonant energy transfer for achieving red afterglow in an aqueous solution. *ACS Nano* **2021**, *15*, 16242–16254.
- [24] Sk, B.; Tsuru, R.; Hayashi, K.; Hirata, S. Selective triplet-singlet Förster-resonance energy transfer for bright red afterglow emission. *Adv. Funct. Mater.* **2023**, *33*, 2211604.
- [25] Zhao, W. J.; He, Z. K.; Tang, B. Z. Room-temperature phosphorescence from organic aggregates. *Nat. Rev. Mater.* **2020**, *5*, 869–885.
- [26] Li, Q. J.; Zhou, M.; Yang, Q. F.; Wu, Q.; Shi, J.; Gong, A. H.; Yang, M. Y. Efficient room-temperature phosphorescence from nitrogen-doped carbon dots in composite matrices. *Chem. Mater.* **2016**, *28*, 8221–8227.
- [27] Jiang, K.; Wang, Y. H.; Cai, C. Z.; Lin, H. W. Conversion of carbon dots from fluorescence to ultralong room-temperature phosphorescence by heating for security applications. *Adv. Mater.* **2018**, *30*, 1800783.
- [28] Peng, H.; Xie, G. Z.; Cao, Y.; Zhang, L. Y.; Yan, X.; Zhang, X.; Miao, S. H.; Tao, Y.; Li, H. H.; Zheng, C. et al. On-demand modulating afterglow color of water-soluble polymers through phosphorescence FRET for multicolor security printing. *Sci. Adv.* **2022**, *8*, eabk2925.
- [29] Xiong, S. D.; Xiong, Y.; Wang, D. L.; Pan, Y. W.; Chen, K. Y.; Zhao, Z.; Wang, D.; Tang, B. Z. Achieving tunable organic afterglow and UV-irradiation-responsive ultralong room-temperature phosphorescence from pyridine-substituted triphenylamine derivatives. *Adv. Mater.* **2023**, *35*, 2301874.
- [30] Sun, W. X.; Zhang, Y. Q.; Yin, G. C.; Lu, S. Y. Afterglow of dual-confined carbon dots ranging from blue to near-infrared. *Adv. Funct. Mater.* **2024**, *34*, 2402346.
- [31] Xu, J. H.; Liang, Q. J.; Li, Z. J.; Osipov, V. Y.; Lin, Y. J.; Ge, B. H.; Xu, Q.; Zhu, J. F.; Bi, H. Rational synthesis of solid-state ultraviolet B emitting carbon dots via acetic acid-promoted fractions of sp³ bonding strategy. *Adv. Mater.* **2022**, *34*, 2200011.
- [32] Xu, Y. L.; Wang, B. Y.; Zhang, M. M.; Zhang, J. X.; Li, Y. D.; Jia, P. J.; Zhang, H.; Duan, L. L.; Li, Y.; Li, Y. T. et al. Carbon dots as a potential therapeutic agent for the treatment of cancer-related anemia. *Adv. Mater.* **2022**, *34*, 2200905.
- [33] Qu, S. N.; Zhou, D.; Li, D.; Ji, W. Y.; Jing, P. T.; Han, D.; Liu, L.; Zeng, H. B.; Shen, D. Z. Toward efficient orange emissive carbon nanodots through conjugated sp³-domain controlling and surface charges engineering. *Adv. Mater.* **2016**, *28*, 3516–3521.
- [34] Chen, Y. H.; Zheng, M. T.; Xiao, Y.; Dong, H. W.; Zhang, H. R.; Zhuang, J. L.; Hu, H.; Lei, B. F.; Liu, Y. L. A self-quenching-resistant carbon-dot powder with tunable solid-state fluorescence and construction of dual-fluorescence morphologies for white light-emission. *Adv. Mater.* **2016**, *28*, 312–318.
- [35] Zhan, Y.; Shang, B.; Chen, M.; Wu, L. M. One-step synthesis of silica-coated carbon dots with controllable solid-state fluorescence for white light-emitting diodes. *Small* **2019**, *15*, 1901161.
- [36] Miao, X.; Qu, D.; Yang, D. X.; Nie, B.; Zhao, Y. K.; Fan, H. Y.; Sun, Z. C. Synthesis of carbon dots with multiple color emission by controlled graphitization and surface functionalization. *Adv. Mater.* **2018**, *30*, 1704740.
- [37] Sun, C.; Zhang, Y.; Ruan, C.; Yin, C. Y.; Wang, X. Y.; Wang, Y. D.; Yu, W. W. Efficient and stable white LEDs with silica-coated

- inorganic perovskite quantum dots. *Adv. Mater.* **2016**, *28*, 10088–10094.
- [38] Zhu, J. Y.; Li, C.; Zhu, Y. S.; Hu, J. H.; Nan, Y.; Chen, X.; Liu, K. K.; Wang, H. L.; Shan, C. X.; Xu, W. et al. Long-wavelength afterglow emission with nearly 100% efficiency through space-confined energy transfer in organic-carbon dot hybrid. *Nano Lett.* **2024**, *24*, 13307–13314.
- [39] Shi, H. X.; Niu, Z. J.; Wang, H.; Ye, W. P.; Xi, K.; Huang, X.; Wang, H. L.; Liu, Y. F.; Lin, H. W.; Shi, H. F. et al. Endowing matrix-free carbon dots with color-tunable ultralong phosphorescence by self-doping. *Chem. Sci.* **2022**, *13*, 4406–4412.
- [40] Zheng, Y.; Zhou, Q.; Yang, Y.; Chen, X. H.; Wang, C.; Zheng, X.; Gao, L.; Yang, C. L. Full-color long-lived room temperature phosphorescence in aqueous environment. *Small* **2022**, *18*, 2201223.
- [41] Shi, M. C.; Gao, Q.; Rao, J.; Lv, Z. W.; Chen, M. X.; Chen, G. G.; Bian, J.; Ren, J. L.; Lü, B. Z.; Peng, F. Confinement-modulated clusterization-triggered time-dependent phosphorescence color from xylan-carbonized polymer dots. *J. Am. Chem. Soc.* **2024**, *146*, 1294–1304.
- [42] Cui, M. Y.; Li, M. J.; Wang, J. H.; Chen, R. Z.; Xu, Z. J.; Wang, J. Y.; Han, J. F.; Hu, G. Y.; Sun, R.; Jiang, X. et al. Hydrothermal synthesis of zinc-doped silica nanospheres simultaneously featuring stable fluorescence and long-lived room-temperature phosphorescence. *Angew. Chem., Int. Ed.* **2021**, *60*, 15490–15496.



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