

A Journal of the Gesellschaft Deutscher Chemiker A Deutscher Chemiker CDCh International Edition Www.angewandte.org

Accepted Article

- **Title:** Full-spectrum persistent luminescence tuning using all-inorganic perovskite quantum dots
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This manuscript has been accepted after peer review and appears as an Accepted Article online prior to editing, proofing, and formal publication of the final Version of Record (VoR). This work is currently citable by using the Digital Object Identifier (DOI) given below. The VoR will be published online in Early View as soon as possible and may be different to this Accepted Article as a result of editing. Readers should obtain the VoR from the journal website shown below when it is published to ensure accuracy of information. The authors are responsible for the content of this Accepted Article.

To be cited as: Angew. Chem. Int. Ed. 10.1002/anie.201901045 Angew. Chem. 10.1002/ange.201901045

Link to VoR: http://dx.doi.org/10.1002/anie.201901045 http://dx.doi.org/10.1002/ange.201901045

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Full-spectrum persistent luminescence tuning using all-inorganic perovskite quantum dots

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Abstract: The applications of persistent luminescence phosphors as night or dark-light vision materials in many technological fields have fuelled up a growing demand for rational control over the emission profiles of the phosphors, which, however, remains a daunting challenge so far. Herein, we report a unique strategy to fine-tuning the persistent luminescence by using all-inorganic $CsPbX_3$ (X = Cl, Br, and I) perovskite quantum dots (PeQDs) as efficient light conversion materials. Full-spectrum persistent luminescence with wavelengths covering the entire visible spectral region is achieved through tailoring of the PeQDs bandgap, in parallel with narrow bandwidth of PeQDs and highly synchronized afterglow decay due to the single energy storage source. These findings break through the limitations of traditional afterglow phosphors, thereby opening up new opportunities for persistent luminescence materials towards many revolutionary applications such as making a white-emitting persistent light source and dark-light multicolor displays.

Persistent luminescence, known also as long-lasting phosphorescence or afterglow, is an optical phenomenon whereby light emission from a material can last for a long time after the cease of excitation. The first scientific description of persistent luminescence dated back to 1602 for the discovery of Bologna stone,^[1] whereas a substantial breakthrough has been accomplished until 1996 when Matsuzawa et al. realized an ultralong green afterglow over 30 h in SrAl₂O₄:Eu²⁺, Dy³⁺ phosphor.^[2] This pioneering work has stimulated a rapid development of persistent luminescence materials in the past two decades.^[3] To date, the blue and green persistent luminescence phosphors have been well developed and commercialized in the markets of luminous paints, safety signs and toys; however, the red persistent luminescence materials have confronted deficiencies of weak persistent luminescence and short afterglow time, though endeavors are still

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- [**] This work is supported by the Strategic Priority Research Program of the CAS (XDB2000000), the CAS/SAFEA International Partnership Program for Creative Research Teams the NSFC (Nos. U1805252, 11774345, 21875250, and 51872048), the Youth Innovation Promotion Association (No. 2016277), the Chunmiao Project of Haixi Institutes of the CAS (No. CMZX-2016-002).
 - Supporting information for this article is available on the WWW under http://www.angewandte.org or from the author.

ongoing.^[4] It is generally accepted that the afterglow performance of persistent luminescence phosphors are determined by the electron (or hole) trap depths and densities in the host.^[5] Particularly, the afterglow time of different persistent luminescence phosphors would vary drastically due to distinct electron (or hole) trap depths. The unbalanced duration, the deficiencies in red component as well as the broad afterglow band of persistent luminescence phosphors have delayed or even precluded many novel applications, including white-emitting persistent light source, dark-light multicolor displays, and ultrasensitive biosensing.^[6] To change this unfavorable status quo, it could be a smart choice to search for an efficient light conversion material to modulate the afterglow emission colors of the phosphors, which, however, remains notoriously difficult so far.

All-inorganic cesium lead halide (CsPbX₃, X = Cl, Br, and I) perovskite quantum dots (PeQDs) have recently evoked considerable attention in the fields of solar cells, light-emitting diodes, lasing and photodetectors, owing to their outstanding optoelectronic properties such as long carrier diffusion distance, large absorption coefficient, high photoluminescence (PL) quantum yield (QY), narrow emission band, and tunable bandgap and PL emission.^[7] Specifically, the PL emission of CsPbX₃ PeQDs can be facilely tuned over the entire visible spectral region by adjusting the halide composition.^[8] These superior features make CsPbX₃ PeQDs ideal candidates as light harvesting and light emitting materials for a variety of optoelectronic and photovoltaic applications.^[9]

Herein, we report a convenient and versatile approach to fine-tuning the persistent luminescence by using CsPbX₃ PeQDs as efficient light conversion materials. In our design (Fig.1), CaAl₂O₄:1%Eu²⁺, 0.5%Nd³⁺ (CAO) afterglow phosphor is embedded in a polydimethylsiloxane (PDMS) substrate and spin-coated onto a quartz plate to function as a light storage layer to harvest the ultraviolet (UV) light and store the excitation energy, which is subsequently released slowly and yields violet and blue afterglow emission. The persistently emitted photons from CAO are then absorbed by CsPbX₃ PeQDs in the light conversion layer, creating electron-hole pairs (excitons) in the conduction band (CB) and valence band (VB) of PeQDs, followed by photon emission through exciton recombination. As a consequence, tunable persistent luminescence with wavelengths covering the entire visible spectral region could be realized via bandgap tailoring of CsPbX₃ PeQDs, in parallel with the benefits of narrow bandwidth of PeQDs and high synchronization in afterglow decays due to the single energy storage source of CAO.



Figure 1. Schematic illustration for full-spectrum persistent luminescence tuning using all-inorganic CsPbX₃ PeQDs.



Figure 2. a) Schematic illustration for the synthesis of CsPbX₃ PeQDs: HX was injected into the hot solution containing Cs⁺ and Pb²⁺ precursors, oleic acid (OA), oleylamine (OAm), trioctylphosphine (TOP) and 1-octadecene (ODE), followed by rapid cooling in ice-water bath. TEM images of the as-synthesized b) CsPbCl₃, c) CsPbCl_{1.5}Br_{1.5}, d) CsPbBr₃, e) CsPbBr₂I, and f) CsPbI₃ PeQDs. The scale bars in (b-f) represent 50 nm, and the insets show the corresponding PL photographs (λ_{ex} = 365 nm) for the colloidal cyclohexane solution of the PeQDs at a concentration of 1 mg mL⁻¹. g) Optical absorption spectra, PL emission spectra (λ_{ex} = 365 nm), and h) PL decays of CsPbX₃ PeQDs with varying halide compositions.

High-quality CsPbX₃ PeQDs were synthesized through a unique hot-injection method (Fig. 2a). The as-prepared PeQDs displayed remarkably bright PL with full-gamut color tuning from violet to deep red, under UV excitation at 365 nm (Insets of Fig. 2b-f). Structure and morphology characterizations through TEM, powder XRD, energy dispersive X-ray spectra, and selected area electron diffraction revealed cubic phase and high crystallinity of the PeQDs with average lengths of 11.2–16.8 nm (Fig. 2b–f and Fig. S1 and S2). Optical absorption spectra showed that the PeQDs had large absorbance in the UV and visible regions with band edges shifting from 408 nm to 692 nm as the halide composition changed from Cl to I. PL emission spectra of the PeQDs exhibited tunable exciton emission bands over the entire visible spectral region, with full widths at half maximum of 9-31 nm (Fig. 2g and Fig. S3). The corresponding PL decays indicated effective PL lifetimes of 7.7-103.1 ns with faster decays for wider bandgap PeQDs (Fig. 2h and Table S1). The absolute PLQYs, defined as the ratio of the number of emitted photons to the number of absorbed photons, were determined to be 38.5-99.0%, with the highest value approaching unity for CsPbI₃ PeQDs (Table S1).

We selected CAO as the sensitizer in view of its excellent persistent luminescence in the violet and blue regions which overlap well with the absorption of most CsPbX₃ PeQDs.^[10] CAO was synthesized through a solid-state reaction method followed by ball milling treatment to yield submicron particles (Fig. S4 and S5). As a result, the CAO submicrons can be mixed homogeneously with SYLGARD 184 silicone elastomer and then spin-coated onto a quartz plate, forming a semi-transparent CAO/PDMS composite film with a thickness of approximately 0.5 mm. Thereafter, a 0.5-mm thick PDMS/CsPbX₃ PeQDs layer was spin-coated onto the surface of CAO/PDMS with a CAO/PeQDs mole ratio of 20:1,



Figure 3. a) Photographs of CAO/PeQDs/PDMS films with varying halide compositions of CsPbX₃ PeQDs under daylight. b) Afterglow images of the films in (a) recorded at a delay time of 10 min after 5-min of illumination with a 365-nm UV lamp. The scale bars in (a, b) represent 1 cm. c) Normalized afterglow emission spectra and d) afterglow decay curves of CAO/PeQDs/PDMS films with varying PeQDs compositions. The afterglow emission spectra and afterglow decay curves of the films were recorded at room temperature with delay times of 10 min and 10 s, respectively, after 5-min of illumination with a 365-nm UV lamp. e) CIE chromaticity diagram showing the color gamut of the afterglow emission colors (solid white triangle) from CAO/PeQDs/PDMS films, compared with the color gamut (dashed white triangle) defined in the NTSC television color standard.

resulting in a series of deeply colored CAO/PeQDs/PDMS composite films (Fig. 3a). The PDMS matrix can also to some extent protect PeQDs from degradation and thus improve their photo-, thermal-, and air-stability (Fig. S6). After irradiation with a 365-nm UV lamp for 5 min, these films displayed bright persistent luminescence with tunable emission colors from purple to green and deep red as the halide composition of the PeQDs changed from Cl to Br and I (Fig. 3b), which remained clearly visible by naked eye even after 3 h of decay (Fig. S7). Afterglow emission spectra of the films showed tunable emission bands from 440 nm to 694 nm (Fig. 3c), which agreed well with their corresponding steady-state PL emission spectra (Fig. S8). This suggests that the persistent luminescence of CAO/PeQDs/PDMS originates from exciton emissions of PeQDs via energy transfer from CAO since PeQDs alone show no afterglow luminescence. Further afterglow decay measurement showed that the afterglow intensities of the films decreased slowly over a long time window and remained considerably high after 8 h of decay (Fig. 3d and Fig. S9), confirming the long afterglow time of the composite films. Moreover, it was found that the afterglow intensities of PeQDs in CAO/PeQDs/PDMS were comparable to that of CAO in CAO/PDMS at equivalent decay times, due to very efficient energy transfer from CAO to PeQDs. Importantly, because of the narrow emission bandwidth of PeQDs, highly saturated RGB afterglow colors can be generated, leading to a wide color gamut encompassing 130% of that defined in the NTSC color standard (Fig. 3e and Table S2). Such a unique afterglow feature combined with narrow bandwidth, wide color gamut and highly synchronized afterglow decay from CAO/PeQDs/PDMS composites is otherwise inaccessible by traditional afterglow phosphors, which is highly desirable for optical coding and dark-light multicolor displays.

To gain more insights into the energy transfer processes in CAO/PeQDs/PDMS composite films, we recorded PeQDs content-dependent afterglow emission spectra. As shown in Fig. 4a and 4b, the afterglow emissions of CsPbCl₂Br at 441 nm and CsPbBr_{1.5}I_{1.5} at 620 nm increased gradually at the expense of that of CAO with increasing contents of CsPbCl₂Br and CsPbBr_{1.5}I_{1.5} PeQDs in the composite films, as a result of energy transfer from CAO to CsPbCl₂Br and CsPbBr_{1.5}I_{1.5}, respectively. Similar



Normalized afterglow emission spectra Figure 4. of a) CAO/CsPbCl₂Br/PDMS and b) CAO/CsPbBr_{1.5}I_{1.5}/PDMS composite films with a fixed CAO content of 100 µmol and varying CsPbCl₂Br and CsPbBr_{1.5}I_{1.5} contents, respectively. c) 100 K PL decays from 4f⁶5d¹ of Eu²⁺ in CAO/PeQDs/PDMS films with CAO/PeQDs molar ratios of 5:1 and different PeQDs compositions by monitoring Eu²⁺ emission at 440 nm. d) TL glow curves of CAO/PDMS, CAO/CsPbCI_{1.5}Br_{1.5}/PDMS, CAO/CsPbBr₃/PDMS, and CAO/CsPbBr_{1.5}I_{1.5}/PDMS by monitoring their afterglow emissions at 440, 468, 523, and 620 nm, respectively. e) Schematic illustration of the proposed persistent luminescence mechanism in CAO/PeQDs/PDMS. The solid-line arrows denote electronic transitions, and the solid-curve and dash-curve arrows represent the trapping and de-trapping of electrons, respectively.

evolutions in PeQDs content-dependent afterglow emissions were also observed for PeQDs with different halide compositions (Fig. S10-S13), which enables us to fine-tune the persistent luminescence of CAO/PeQDs/PDMS films by simply adjusting the contents of PeQDs. It is noteworthy that, the afterglow band of Eu²⁺ from CAO in CAO/CsPbCl2Br/PDMS was selectively quenched in accordance with CsPbCl₂Br absorption (Fig. S14): the short-wavelength side (400-441 nm) of the afterglow band declined gradually with increasing CsPbCl2Br content, whereas its long-wavelength side (441-520 nm) remained nearly unchanged (Fig. 4a). One should keep in mind that the depopulation of 4f65d1 of Eu2+ through nonradiative Förster resonance energy transfer (FRET) would lead to simultaneous quenching of the whole emission band of 4f⁶5d¹ of Eu^{2+} . Therefore, the selective quenching in the afterglow band of Eu²⁺ in CAO/CsPbCl₂Br/PDMS with increasing CsPbCl₂Br content indicates that the CAO-to-CsPbCl2Br energy transfer results from a radiative reabsorption process instead of a nonradiative FRET process. Such radiative energy transfer from CAO to PeQDs can be further verified by comparing the PL lifetimes of Eu2+ in CAO/PeQDs/PDMS composites of different PeQDs contents and compositions, since nonradiative FRET would otherwise result in a decrease in PL lifetime of Eu²⁺ by imposing additional relaxation channel on the 4f65d1 excited state of Eu2+.[11] To avoid the interference from strong afterglow signal at room temperature, we measured the PL lifetimes of the composites at 100 K. It turned out that the PL lifetimes of Eu^{2+} in the composites were essentially identical (587 ns) and independent of the contents and compositions of PeODs (Fig. 4c and Fig. S15), thus demonstrating unambiguously that the CAO-to-PeQDs energy transfer is dictated by a radiative reabsorption process.

In addition to probing the energy transfer processes, we also survey the trapping and de-trapping processes underlying the persistent luminescence of CAO/PeQDs/PDMS composite films. Figure 4d compares the thermoluminescence (TL) glow curves of CAO/PDMS and CAO/PeQDs/PDMS with different PeQD compositions. It was found that the TL glow curves of PeQDs in



Figure 5. Afterglow emission spectra for the mixture of a) CAO, CsPbBr₃ and KSF, and b) CSAO, YAG and CsPbBr_{1.5}I_{1.5}, measured at a delay time of 10 min after the samples were illuminated with a 365-nm UV lamp for 5 min. The insets show the corresponding afterglow images of the mixed powder. c) Multicolor persistent luminescence imaging (left, bright-field image; right, afterglow images recorded at a delay time of 10 min after 5-min of illumination with a 365-nm UV lamp) by casting CAO and CsPbX₃ PeQDs of different compositions onto the flexible PDMS substrates (purple: CAO; blue: CsPbCl_{1.5}Br_{1.5}; green: CsPbBr₃; dark yellow: CsPbBr_{1.8}I_{1.2}; red: CsPbBr_{1.5}I_{1.5}). The scale bar represents 2 cm.

CAO/PeQDs/PDMS were generally consistent with that of CAO, with nearly identical TL glow peaks at around 360 K. The unchanged TL glow essentially performance between CAO/PeQDs/PDMS and CAO/PDMS demonstrates unequivocally that CAO dictates the energy storage and release processes for the persistent luminescence of CAO/PeQDs/PDMS. By fitting the TL glow curve with the peak-shape method developed by Chen *et al.*,^[12] the dominant trap depth (E_t) in CAO was calculated to be 0.50 eV (Fig. S16), which accounts for the long-lasting phosphorescence of CAO at room temperature.^[13] Based on the above analyses, we propose a physical model to unveil the persistent luminescence mechanism of PeQDs in CAO/PeQDs/PDMS (Fig. 4e). In this model, the traps are supposed to be electron traps lying below the conduction band minimum (CBM) of CAO with their nature ascribed to the lattice defects intrinsic to CAO such as oxygen vacancies (V₀²⁺). Nd³⁺ ion does not directly participate in the electron trapping process but interacts with the oxygen vacancies and increases the trap depth.^[14] Upon UV irradiation, the electrons on the 4f levels of Eu^{2+} can be excited to the 5d levels which are slightly below the CBM of CAO.[15] Then, partial electrons in the 5d levels of Eu²⁺ return to the 4f levels and yield PL emission, and the rest of excited electrons transfer to the CB and are trapped by the empty defect levels, resulting in oxidation of Eu²⁺ into Eu³⁺.^[16] The trapped electrons can then move back to the CB, be captured by Eu³⁺ and populated the 5d levels of Eu²⁺ through thermal activation, accompanied with light emission through $4f^{6}5d^{1} \rightarrow 4f^{7}$ transitions of Eu²⁺.^[17] This process is very slow due to the deep traps produced by Nd³⁺ dopants, which leads to the long afterglow luminescence.^[18] The continuously emitted photons from CAO are subsequently absorbed by PeQDs, creating excitons in PeQDs followed by photon emission from PeQDs through exciton recombination.

A key feature of efficient afterglow light conversion in CAO/PeQDs composites is that the PeQDs possess large absorption coefficient and high PLQY. In this regard, such afterglow light conversion strategy is versatile and can be extended to other energy

storage and light conversion materials. For example, by mixing CAO with the commercial K₂SiF₆:Mn⁴⁺ (KSF) phosphor,^[19] red persistent luminescence was obtained (Fig. S17). Likewise, yellow or red persistent luminescence was achieved by blending the commercial $Ca_{0.9}Sr_{0.1}Al_2O_4:Eu^{2+},Dy^{3+}$ (CSAO) blue afterglow phosphor with Y₃Al₅O₁₂:Ce³⁺ (YAG) phosphor or CsPbBr_{1.5}I_{1.5} PeQDs, respectively (Fig. S18).^[20] As a result, white-emitting persistent light is possible by combining individual blue, green (or yellow), and red afterglow components through light conversion. In the proof-of-concept experiment, we combined the green-emitting CsPbBr3 PeQDs and red-emitting KSF phosphor with the blue-emitting CAO afterglow phosphor. The corresponding afterglow emission spectrum showed characteristic emission bands of CAO, CsPbBr3 and KSF at 440, 523 and 630 nm, respectively (Fig. 5a), with color coordinates at (0.318, 0.309) in CIE 1931. Similarly, white-emitting persistent luminescence with color coordinates at (0.324, 0.349) was derived by combining the vellow-emitting YAG phosphor (560 nm) and red-emitting CsPbBr₁₅I₁₅ PeODs (620 nm) with the blue-emitting CSAO (468 nm) (Fig. 5b), despite its currently low persistent luminescence efficiency due mainly to light scattering under non-optimized conditions. Furthermore, we revealed the potential of the CAO/PeQDs composite materials for dark-light multicolor displays by casting CAO and CsPbX₃ PeQDs of different compositions onto the flexible PDMS substrates. As shown in Fig. 5c, by empolying CsPbCl1.5Br1.5, CsPbBr3, and CsPbBr1.5I1.5 PeQDs for afterglow light conversion, blue, green, and red afterglow images with designed patterns such as the logo of an institute, fish, and flower petal were acquired.

In summary, we have realized full-spectrum persistent luminescence tuning with a color gamut of 130% NTSC and highly synchronized afterglow decay in CAO/CsPbX₃ PeQDs composites by using PeQDs as efficient light conversion materials. These findings provide a general approach for fine-tuning the persistent luminescence of afterglow phosphors, which may open up new opportunities for persistent luminescence materials towards many emerging applications such as fabricating a white-emitting persistent light source for energy-saving thruway lighting, dark-light multicolor displays in billboard, and afterglow coding for complex data storage or multilevel anticounterfeiting. Future efforts on these aspects lie in the development of high-performance UV-emitting afterglow phosphors which can effectively sensitize PeQDs, the optimization of the composite structure to improve the light conversion efficiency, and the exploration of lead-free PeQDs with high stability and PL efficiency.

Experimental Section

Detailed experimental procedures are reported in the Supporting Information.

Received: ((will be filled in by the editorial staff)) Published online on ((will be filled in by the editorial staff))

Keywords: Perovskite, quantum dot, persistent luminescence, energy transfer.

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Table of Contents

Perovskite Quantum Dots

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Full-spectrum persistent luminescence tuning using all-inorganic perovskite quantum dots



Full-spectrum persistent luminescence tuning: A unique and versatitle strategy was proposed to fine-tuning the persistent luminescence over the entire visible spectral region with narrow bandwidth and highly synchronized afterglow decay by using $CaAl_2O_4$:Eu²⁺,Nd³⁺ (CAO) afterglow phosphor as the single energy storage source and all-inorganic CsPbX₃ (X = Cl, Br, and I) perovskite quantum dots (PeQDs) as efficient light conversion materials.