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Design of 2D Templating Molecules for Mixed-Dimensional Perovskite Light-Emitting Diodes

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ABSTRACT: Recent advances in Ruddlesden Popper (RP) perovskites have shown highly efficient light-emitting diodes (LEDs) due to the energy confinement by an energy cascade from the quasi-2D (donors) to 3D (acceptors) states. This is achieved by forming mixed-dimensional phases using 2D templating (spacer) molecules. The effect of the spacer molecular design on the efficacy of light emission is hitherto unknown, which triggered this study into eight different spacer molecules ranging from the naphthyl to phenyl alkyl families. The phase distribution can be modulated by the choice of this molecule alone, which affects the energy landscape. It was found that a high acceptor-to-donor ratio is required to achieve the best LED efficiencies. A simple absorption spectroscopy measurement can also be employed to screen potential RP systems. The findings are of significance to other halide perovskites and lay down a useful guideline to assess new spacer molecules for more efficient RP perovskite LEDs.

1. INTRODUCTION

The achievement of high power conversion efficiencies in halide perovskite solar cells has driven research and development into a wider field of optoelectronics. The past decade has seen remarkable contributions and scientific advances from research groups all over the world, not just in photovoltaic applications¹⁻⁷ but also in light-emitting devices and beyond.⁸⁻¹⁵ In recent years, there have been increasing interests in lower and mixed-dimensional perovskites, also known as Ruddlesden-Popper (RP) perovskites, due to their superior optical and electronic properties enabling more efficient light emission.¹⁶⁻¹⁹ RP perovskites have a general formula of $L_nA_{n-1}B_nX_{3n+1}$, where A, B and X are the constituents of the 3D perovskite, L is a bulky ammonium cation inserted within the 3D perovskite framework forming layered structures, and n represents the number of dimensionally confined BX_6 octahedra layers.²⁰ In this case, $n = 1$ denotes a pure 2D perovskite (L_2BX_4), $n = \infty$ denotes a pure 3D perovskite (ABX_3), and $1 < n < \infty$ represents a quasi-2D mixed-dimensional perovskite (Figure 1a). 2D perovskites have shown promising advantages over their 3D counterpart, such as high exciton binding energies,²¹⁻²³ highly crystalline thin

films with smooth morphologies,²⁴⁻²⁵ and better resistance against moisture, heat and light-induced degradation,²⁶⁻²⁸ although the isolated octahedral layers mean poor charge transport thus limiting their employment in electronic devices.²⁹⁻³¹

By combining both 3D and 2D perovskites to form mixed-dimensional RP phases, benefits of charge transport and enhanced radiative recombination can both be achieved, leading to higher photoluminescence (PL) quantum yield (PLQY) and device performance in these mixed systems.^{29, 32-34} In addition to surface defect passivation,³⁵ the co-existence of multi-phases in the film triggers an energy funneling phenomenon as a result of the different band gaps of the different RP phases.³⁶⁻³⁸ Here, the higher band gap domains from the lower dimensional phases act as donors while the lowest band gap domain from the 3D phase acts as acceptors. The energy transfer from the donor to the acceptor states leads to a high concentration of carriers in the acceptor domains. This cascading energy landscape is particularly beneficial in LEDs due to the enhanced energy confinement which promotes higher radiative recombination that largely outcompetes non-radiative losses.

Various studies utilizing different 2D templating molecules such as butylammonium (BA),³⁹⁻⁴¹ phenylethylammonium (PEA),⁴²⁻⁴⁶ and many others⁴⁷⁻⁵¹ have all shown enhanced photo- and electroluminescence (EL) properties as compared to the pristine system, indicating a promising prospect in this direction. However, most literatures primarily focus on only one specific type of molecule while varying its acceptor-to-donor ratio, an approach adopted in our previous work with PEA.⁵² The effect of the choice of the 2D templating molecules on the efficacy of light emission remains unexplored, which presents a knowledge barrier when it comes to selecting the type of molecules for RP perovskites. The different energy landscape created by changing the spacer molecule is expected to have an effect on both the film properties and device performances, and uncovering this missing information will be important in designing more efficient mixed-dimensional systems for light emission.

In this work, we have explored and compared RP perovskites based on different 2D templating molecules from the naphthyl and phenyl alkyl families (Figure 1b), with the 3D component fixed as CsPbBr₃. Since the concentration of the 2D templating molecule is an important factor that determines the performance of the RP perovskites, different concentrations of each molecule are added to ensure that the comparison across the different systems is done for the best performing composition of each system. The properties of these films are evaluated, along with the corresponding PeLED devices fabricated and tested. Here, we found that the phase distribution of the RP perovskites greatly depend on the spacer structure, which then have a direct impact on the device performance. By comparing across similar 2D templating molecules at different concentrations, we observe that the energy landscape should be tailored towards a high acceptor-to-donor ratio in the film since the acceptor states are where energy is funnelled to and where recombination and light emission takes place, and this is important in determining the efficiency of the EL from the RP devices. In doing so, we have achieved RP PeLEDs with an external quantum efficiency (EQE) of 6%, almost 2 orders of magnitude higher than the 3D device. We also highlight that future screening of new 2D templating molecules can be done easily by monitoring the phase distribution of the RP perovskites through the use of simple absorption spectroscopy techniques.

2. RESULTS AND DISCUSSION

We first investigated mixed-dimensional CsPbBr₃ perovskites based on the naphthyl alkyl ammonium family of molecules, while the phenyl alkyl family is used to substantiate the findings later on. Due to the asymmetry of the naphthyl molecules, we also included possible regioisomers to derive four different naphthyl molecules listed as such: 1-naphthylmethylammonium bromide (1-NMA), 2-naphthylmethylammonium bromide (2-NMA), 1-naphthylethylammonium bromide (1-NEA), and 2-naphthylethylammonium bromide (2-NEA) (molecular

structures shown in Figure 1b). The synthesis procedure can be found in Section S1.1 of the Supporting Information. To verify the formation of 2D perovskites with the NMA and NEA molecules, we first dissolved each 2D templating molecule (L) with PbBr₂ in the composition L₂PbBr₄. The solutions were spin coated and characterized with X-ray diffraction (XRD), with the diffraction patterns shown in Figure 1c. The XRD patterns show regular 2-theta spacings for all four naphthyl-based perovskites, which is characteristic of 2D perovskites. The diffraction peaks of 2-NMA and 2-NEA also match those reported in literature.⁵³ By incorporating the 2-theta values of the diffraction peaks into Bragg's Law, the d-spacing for the (001) plane of the 2D perovskites are calculated as: 1-NMA (2.07 nm), 2-NMA (1.88 nm), 1-NEA (2.05 nm), and 2-NEA (1.97 nm). All four 2D naphthyl perovskites exhibit strong excitonic absorption behaviour as shown in Figure S2 and S3, which is also characteristic of 2D perovskites due to their high exciton binding energy.

The naphthyl molecules are added to the CsPbBr₃ precursors at various concentrations to form RP perovskites. From the field emission scanning electron microscopy (FESEM) images (Figure S4), the RP films are more compact as compared to the pristine CsPbBr₃, similar to that reported previously.⁵² A fully covered and pinhole-free film morphology can be achieved with an excess concentration of ~ 40 to 60 mol%, along with reduction in grain size. The deposited films with different concentrations of naphthyl molecules under UV light are shown in Figure S5, showing highly luminescent green films as compared to the pristine CsPbBr₃, although at higher concentrations of 100 mol%, 2-NMA, 1-NEA and 2-NEA films appear more blueshifted. The phase distribution of the RP film is highly important as it determines the energy landscape and amount of emitting sites,³⁷ which then affects the funneling process and radiative recombination. To investigate the distribution of the RP phases in our naphthyl-based systems, UV-vis absorption spectroscopy was employed (Figure 2). Here, it is noticed that the formation of quasi-2D phases varies with different 2D templating molecules used. While 1-NMA shows only one obvious excitonic peak at 392 nm (Figure 2a), which corresponds to the absorption of the n=1 phase, the other three samples show multiple excitonic peaks corresponding to the n=1, 2, and 3 phases (Figure 2b-d). More importantly, it is observed that the 3D absorption component for the 1-NMA samples is more retained with increasing 1-NMA concentration from 40 to 100 mol%, where the slope at 525 nm remains sharp despite a reduction in the intensity. All three other naphthyl samples, however, saw a huge diminution of the 3D absorption peak with increasing concentration of 2D templating molecules, along with the formation of more quasi-2D phases, implying that there is a lower acceptor-to-donor ratio for these films. This means that the amount of 3D domains where energy can funnel into and undergo recombination is limited. This observation is more clearly distinguished by plotting the same concentration of each molecule in the same graph as shown in Figure S6. Here, it is evidently seen that for all concentrations, the 1-NMA

samples always retain the highest 3D component (black curves), implying that the phase in these 1-NMA films is still dominated by a high acceptor-to-donor ratio.

We next explored how the phase distribution affects the photoexcited processes in our RP films using transient absorption (TA) spectroscopy (Figure 3). Due to the weaker excitonic absorption at lower wavelengths for lower spacer molecule concentrations (Figure S7), we have performed TA measurements of all samples at 100 mol% excess 2D templating molecule. Here, the 1-NMA sample (Figure 3a) exhibits a long-lived photobleaching (PB, $\Delta T/T > 0$) peak at the 3D band gap position (~ 515 nm), implying a large exciton population in the acceptor phase. Comparatively, the other three samples exhibit shorter-lived PB peaks which are blueshifted from the 3D absorption wavelength, corresponding to exciton populations in the quasi-2D states (Figure 3b-d). In particular, the 2-NMA sample (Figure 3b) displays only short-lived PB signals in the low n regions (~ 430 and 460 nm), indicating that the film is dominated by low dimensional phases. Meanwhile, both NEA samples show not only low n PB signals, but also a broadened PB peak closer to the 3D band gap, implying a mixed distribution of low and high n species, although the 3D phase is absent. Since the presence of the 3D component plays a key role for exciton localization, recombination and efficient light emission,⁵⁴ we postulate that the phase distribution in our films plays a major role in the LED performance.

We speculate that the differences in the solubility of the 2D templating molecules are responsible for the crystallization dynamics of the system which then determine the phase formation.⁵⁵ From the solubility data of the naphthyl molecules in dimethyl sulfoxide (DMSO) (Figure S9), 1-NMA shows the highest solubility, followed by 1-NEA, 2-NMA and 2-NEA. It is likely that the poor solubility of the 2-NMA and 2-NEA molecules led to the preferential crystallization of the lower dimensional phases during film formation, leading to higher amount of these donor species as shown in the absorption and TA characterizations. As a result of this phase distribution, the PL spectra of the naphthyl-based RP perovskites (Figure S10-11) show a larger blueshift from the main CsPbBr₃ emission peak for the 2-NEA and 2-NMA samples, along with peak broadening which causes the full width half maximum (FWHM) to increase to more than 20 nm (Figure S12). The 1-NMA sample, on the other hand, shows the smallest shift in PL and retains its high colour purity with a PL FWHM of less than 20 nm even at a high excess concentration of 100 mol%. The PLQY results are tabulated in Figure 4. Here, the poor emission of the 3D CsPbBr₃ (PLQY $\sim 10.9\%$) stems from its poor morphology and lack of energy funnelling. On the other hand, the mixed-dimensional systems show improvement in PLQY to over 20%, except 40 mol% 2-NMA, which gave a PLQY of 16%. The higher PLQY for all mixed-dimensional samples is due to the effect of energy funnelling. The highest PLQY of 47.6% is obtained from the 60 mol% 1-NMA sample, which means that for the same photoexcitation, it exhibits the most efficient energy transfer, thus giving the highest amount of emitted photons. This is ascribed to its phase

distribution, where the high acceptor-to-donor ratio allows a more efficient funnelling effect (Figure S14).

To compare the EL properties of the naphthyl-based RP perovskites, PeLED devices based on each naphthyl molecule were fabricated with different excess concentrations from 20 to 100 mol%. The device architecture, current density and luminance (J-V-L) characteristics are shown in Figure S15, with the current efficiency and EQE graphs plotted in Figure S16 and S17 respectively. The characteristics of the best performing PeLED (60 mol% 1-NMA) are shown in Figure 5, while a summary of the characteristic device parameters is tabulated in Table S1. As compared to the 3D CsPbBr₃ device, all four naphthyl-based RP PeLEDs show lower turn-on voltage (V_{th}), higher luminance and efficiencies. Comparing the four different naphthyl-based RP devices, the best concentration lies around 40 or 60 mol%. The 1-NMA device outperforms the other three molecules, with current efficiency and EQE of 20.2 cd A⁻¹ and 6.01% respectively (Figure 5b), which is almost 2 orders of magnitude higher than the 3D CsPbBr₃ (0.27 cd A⁻¹ and 0.07%) (Table S1). The best efficiencies for 2-NMA, 1-NEA and 2-NEA are 3.91 cd A⁻¹ (1.22%), 6.79 cd A⁻¹ (2.45%), and 5.86 cd A⁻¹ (1.98%) respectively. The V_{th} for 1-NMA is lower at 2.6 V as compared to the other three naphthyl devices, which show turn-on voltages of more than 3 V. All four naphthyl PeLEDs show high luminance of close to or even higher than 10 k cd m⁻², as compared to the 3D CsPbBr₃ PeLED, which gave less than 1 k cd m⁻². The device lifetimes (t_{50}) under constant current are measured for the best composition of each mixed-dimensional system, showing the longest lifetime of 22.7 min for the 1-NMA device, 11.4 min for 2-NEA, 7.5 min for 1-NEA, and 5.6 min for 2-NMA (Figure S19).

To assess why the 1-NMA templating molecule performs better than the others, we next investigated the correlation between the phase distribution of the RP perovskites and their corresponding device performances. The efficiencies against additive concentration for each of the naphthyl molecules are plotted in Figure 6a and S15 for a better comparison of the samples at each concentration. As it is speculated that the acceptor-to-donor ratio plays an important role in the emission efficacy in the mixed-dimensional devices, the absorption spectra at each concentration (zoomed in at the 3D absorption onset) are also shown in Figure 6b-d, while the absorbance and EQE trends are plotted in Figure 6e. By comparing the EQE trends to the absorption curves at each concentration, we observe that the efficiency trend corresponds very closely to the proportion of 3D phase. For instance, at 40 mol% excess 2D templating molecules, the higher 3D component of 1-NMA followed by 2-NEA resulted in a similar pattern in the EQE (Figure 6e). This correlation is again observed for the 60 mol% samples. At 100 mol%, all but the 1-NMA sample have their 3D absorption component almost completely diminished, which lead to EQEs close to 0%, whereas the 1-NMA sample still retains a significant amount of 3D absorption component and thus is still able to achieve an EQE of 1.61% at this high concentration. From this comparison, the acceptor-to-donor ratio appears to

have a direct impact on the performance of RP PeLEDs. This is because it sets the energy landscape and determines the amount of low band gap acceptor domains that energy can funnel into and undergo radiative recombination. It is hereby shown that the phase distribution can be modulated by the choice of the 2D templating molecule and should be tailored towards a high acceptor-to-donor ratio in order to achieve higher efficiency devices. In addition, we have verified that the differences in device performances are not due to changes in the electrical properties, which can affect charge balance and injection barrier, but mainly due to the phase distribution that affects the funnelling efficiency (more discussion can be found in Section S1.6, Figures S20–21 in the Supporting Information).

The EL spectra of the naphthyl-based RP PeLEDs are shown in Figure S22, with the FWHM plotted in Figure S23. Likewise to the PL observations, the emission peaks from the 1-NMA devices show the smallest blueshift and a narrow FWHM of 20 nm at 60 mol% concentration. This implies that the colour purity can be retained if the phase distribution is tailored towards a high acceptor-to-donor ratio, which will then enable a wide colour gamut on the Commission internationale de l'éclairage (CIE) colour space (Figure S24).

We tried to extrapolate the findings of the naphthyl-based RP perovskites to the phenyl alkyl systems to verify the trend across other types of spacer structures. In this case, a phenyl group with four different attached carbon chain lengths was used with their molecular structures shown in Figure S26: phenylmethylammonium bromide (PMA), phenylethylammonium bromide (PEA), phenylpropylammonium bromide (PPA), and phenylbutylammonium bromide (PBA) (Solubility information shown in Figure S27). Here, the device performances (Figure S28–29, Table S3) peak at 75 mol% excess 2D templating molecules, showing the highest current efficiency and EQE for PEA (9.07 cd A⁻¹, 2.78%), followed by PPA (6.21 cd A⁻¹, 1.87%), PBA (4.97 cd A⁻¹, 1.70%) and PMA (2.17 cd A⁻¹, 0.69%). The device efficiencies against additive concentration is again plotted in Figure S30 and S31a with the absorption spectra for the 75 mol% samples (zoomed in at the 3D absorption onset) shown in S31b. The absorbance and EQE trends are shown in Figure S32 for a clearer comparison. Likewise, the efficiency trend closely follows the proportion of the absorption from the 3D acceptor states, where the absorption component is highest for PEA followed by PPA. This again reiterates the importance of a high acceptor-to-donor ratio in the phase distribution of these RP perovskites, which depends on the 2D templating molecule used and has a direct impact on the device performance. The simple absorption spectroscopy characterization technique is also shown to be a useful tool in screening of new 2D templating molecules. Finally, the best EQEs of each 2D templating molecule in this work are summarized in Figure 7, while a comparison against other literature work is tabulated in Table S4.

3. CONCLUSIONS

In conclusion, we have explored and studied RP PeLEDs based on eight different 2D templating molecules from the naphthyl and phenyl alkyl families. The choice of this 2D templating molecule is found to be able to tune the phase distribution of the RP phases, which then affects the energy landscape in the film. A phase distribution that is tailored towards a high acceptor-to-donor ratio is vital as this is the main domain where energy is funneled and excitons are localized, and where radiative recombination and light emission occurs. This then translates to the best performing device fabricated with 60 mol% addition of 1-NMA, showing the highest current efficiency and EQE of 20.2 cd A⁻¹ and 6.01% respectively. The efficiency trend of the devices is directly in line with the absorption component of the 3D acceptor states, which is likewise observed in the phenyl alkyl molecular study. This implies that future screening of 2D templating molecules can be easily done through the use of simple UV-vis absorption spectroscopy, making the screening process quick and high-yield. It should be noted that the scattering across different samples, which could be due to varying roughness, needs to be taken into account and should be similar in order to provide a more accurate comparison. The conclusions may not necessarily apply to the inorganic perovskite system alone, but also to the organic-inorganic hybrid counterparts such as methylammonium and formamidinium perovskite systems. Further studies can be done to understand the precise morphology and location of individual RP phases and how these factors can contribute to the overall optical properties. From here, future exploration into new spacer molecules and design of next generation RP perovskite emitters can be made more efficient.

4. EXPERIMENTAL SECTION

Materials: CsBr (99.9%), PbBr₂ (99.999%), 3-phenyl-1-propylamine (98%), 4-phenylbutylamine (98%), 1-(chloromethyl)naphthalene (97.0%), 2-(bromomethyl)naphthalene (96%), hydrobromic acid (48%) and poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) (3.0–4.0% in H₂O) were purchased from Sigma Aldrich. Phenylammonium bromide, benzylammonium bromide and phenylethylammonium bromide were purchased from GreatCell Solar. 1,3,5-Tris(1-phenyl-1Hbenzimidazol-2-yl)benzene (TPBi) (sublimed, > 99.5%) was purchased from Lumtec. All chemicals were used as received, except the naphthyl-based precursors, which require additional synthesis steps to derive the naphthyl alkyl ammonium bromide salts. Detailed synthesis of the naphthyl 2D templating molecules can be found in the Supporting Information Section S1.1.

Perovskite film formation: The CsPbBr₃ perovskite precursor solution was prepared by mixing CsBr and PbBr₂ in the molar ratio of 1.2:1, and dissolved in DMSO to a molar concentration of 0.3 M. The mixed-dimensional compositions were prepared with precise ratios of the 2D templating molecules in excess mol% with respect to

PbBr₂. The mixtures were blended vigorously on a vortex mixer to ensure complete dissolution before using. Thin films were deposited by spin coating the precursor solutions at 5000 rpm for 30 s in an argon-filled glove box. The films were left for 30 min before annealing at 70 °C for 1 min.

PeLED device fabrication: Indium tin oxide (ITO) coated glass substrates (8 Ω sq⁻¹) were pre-cleaned with decon soap, deionized water, acetone and isopropyl alcohol sequentially in a bath sonicator, followed by a UV-ozone treatment for 30 min. PEDOT:PSS was first deposited by spin coating at 4000 rpm for 60 s followed by annealing at 200 °C for 40 s. The substrates were then transferred into an argon-filled glove box for perovskite deposition. Thereafter, TPBi (45 nm), LiF (0.8 nm), and Al (100 nm) were thermally evaporated at a base pressure of < 3 × 10⁻⁶ Torr to complete the device. A shadow mask is used for the electrode deposition and the final active area is 8 mm². The devices were encapsulated in the glove box before testing.

Physical characterization: The absorption spectra were collected using a UV-Vis-NIR spectrophotometer (UV 3600, Shimadzu) equipped with an integrating sphere. The steady-state PL spectra were measured using a Fluoromax-4 (Horiba Jobin Yvon) spectrofluorometer at an excitation wavelength of 350 nm. The PLQY measurement was conducted by placing the sample in an integrating sphere (Labsphere) and exciting it with a 445 nm continuous wave diode laser (Cobolt). The output of the integrating sphere is coupled through an optical fiber to a charge-coupled device (Newton 920 CCD, Andor) in which the photon count is captured. The XRD measurements were performed using a Bruker D8 Advance X-ray diffractometer using Ni-filtered Cu Kα radiation. FESEM imaging of perovskite films (on ITO/PEDOT:PSS coated substrated) was done using a JEOL JSM-7600F scanning electron microscope with an accelerating voltage of 5 kV. The femtosecond TA spectroscopy measurements were performed using the Ultrafast System HELIOS TA spectrometer. The laser source was generated by the Coherent Legend regenerative amplifier (150 fs, 1 KHz, 800 nm) seeded by a Coherent Vitesse oscillator (100 fs, 80 MHz). The broadband probe pulses (450–750 nm) were created by focusing a small portion (around 10 μJ) of the primary 800 nm laser pulses into a 2 mm sapphire plate, while the 400 nm pump pulses were produced by frequency doubling the 800 nm pulses with a BBO crystal.

Device characterization: PeLED devices were tested at ambient conditions in a fully dark environment to eliminate contribution from stray light. The current density and voltage (J-V) characteristics were recorded using a Keithley 2612B source meter while the luminance (L) was collected in an integrating sphere (OceanOptics FOIS-1) coupled to an OceanOptics QE Pro spectrometer. The measurements were operated using a Ciemo LabVIEW software from a computer. An OceanOptics HL-3 Plus vis-NIR LED, calibrated via the ISO 17025, IEC Guide 115 and JCGM100:2008 (GUM) protocols was employed to calibrate the spectrometer's quantification of absolute irradiance.

ASSOCIATED CONTENT

Supporting Information. Materials synthesis, molecular structures, additional comparisons of absorption, TA and PL spectra, FWHM analyses, solubility data, SEM images, J-V-L, current efficiency and EQE characteristics of fabricated PeLED devices, corresponding CIE coordinates. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest.

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Figure 1. (a) Schematic illustration of the different dimensional structures of RP perovskites: (left) $n = \infty$ represents a 3D perovskite; (middle) $1 < n < \infty$ represents a quasi-2D perovskite; (right) $n = 1$ represents a 2D perovskite. (b) Various 2D templating molecules employed in this work. (c) XRD patterns of 2D naphthyl-based perovskites: $(1\text{-NMA})_2\text{PbBr}_4$ (black), $(2\text{-NMA})_2\text{PbBr}_4$ (red), $(1\text{-NEA})_2\text{PbBr}_4$ (blue), and $(2\text{-NEA})_2\text{PbBr}_4$ (magenta).

Figure 2. UV-vis absorption spectra of mixed-dimensional CsPbBr₃ perovskite films with different mol% excess concentration of 2D templating molecules: (a) 1-NMA, (b) 2-NMA, (c) 1-NEA, and (d) 2-NEA.

Figure 3. Transient absorption (TA) spectra ($\Delta T/T$) of mixed-dimensional CsPbBr₃ perovskite films with 100 mol% excess of (a) 1-NMA, (b) 2-NMA, (c) 1-NEA, and (d) 2-NEA at different delay times. The star in (a) shows the desired photobleaching (PB) at the 3D region, while the crosses in (b) and (d) show the undesired PB peaks that correspond to low dimensional phases.

Figure 4. PLQY of mixed-dimensional CsPbBr_3 films with 40 and 60 mol% excess concentration of various naphthyl-based 2D templating molecules.

Figure 5. Device characteristics of the best performing mixed-dimensional PeLED using 60 mol% 1-NMA: (a) J-V-L characteristics, (b) current efficiency and EQE graphs, and (c) PL and EL spectra with the inset showing the corresponding CIE coordinates.

Figure 6. (a) Comparison of EQE of mixed-dimensional CsPbBr₃ PeLEDs with the different naphthyl molecules against concentration. The efficiency trend at each concentration closely follows the 3D phase absorption intensity: (b) 40, (c) 60, and (d) 100 mol% excess concentration. (e) Plot of 3D absorbance and EQE trend of each naphthyl templating molecule at different concentrations.

Figure 7. Summary of the best EQE achieved with various 2D templating molecules used in this work.

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How to select a 2D templating molecule for mixed-dimensional perovskite light-emitting diodes: Effect of the choice of spacer structure is investigated through comparison across eight different molecules, where the importance of phase distribution and acceptor-to-donor ratio in the mixed-dimensional perovskite film is highlighted by a significant enhancement in device efficiencies.