Supplementary Information

Quantum-Cutting Yb^{3+}-Doped Perovskite Nanocrystals for Monolithic Bilayer Luminescent Solar Concentrators

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$CuInS_2$ LSC Layer Modeling

The model described here was developed and validated in a previous report.[1] In this model, the fraction of absorbed photons attenuated by the waveguide, re-absorbed once by the NCs, and emitted into the escape cone are calculated separately using equations s1, s2, and s3, respectively.

$$\mathcal{F} = \frac{1}{4\pi L^2}\iint_{L\times L}\int_0^{2\pi}\int_{\varphi_{esc}}^{\pi-\varphi_{esc}}\int PL_{norm}(\lambda)\frac{\alpha}{\alpha + A_{abs}(\lambda)}\left(1 - e^{-(\alpha - A_{abs}(\lambda))l(x,y,\theta,\varphi)}\right)\sin\varphi\, d\lambda d\varphi d\theta dx dy \quad \text{(s1)}$$

$$\mathcal{R} = \frac{1}{4\pi L^2}\iint_{L\times L}\int_0^{2\pi}\int_{\varphi_{esc}}^{\pi-\varphi_{esc}}\int PL_{norm}(\lambda)\frac{A_{abs}(\lambda)}{\alpha + A_{abs}(\lambda)}\left(1 - e^{-(\alpha - A_{abs}(\lambda))l(x,y,\theta,\varphi)}\right)\sin\varphi\, d\lambda d\varphi d\theta dx dy \quad \text{(s2)}$$

$$\mathcal{E} = 1 - \sqrt{1 - \frac{1}{n^2}} \quad \text{(s3)}$$

Here, α is the attenuation of the waveguide (0.002 cm^{-1}). From here, the proportion of emitted photons that reach the edge of the LSC without being re-absorbed, attenuated, or emitted through the escape cone is $\mathcal{L} = 1 - \mathcal{F} - \mathcal{R} - \mathcal{E}$. To account for multiple cycles of re-absorption and emission, the $OQE(L)$ of the $CuInS_2$ NC layer is calculated using an infinite series as follows:

$$OQE(L) = \mathcal{L}\Phi\sum_{i=0}^{\infty}(\mathcal{R}\Phi)^i = \frac{\mathcal{L}\Phi}{1 - \mathcal{R}\Phi} \quad \text{(s4)}$$

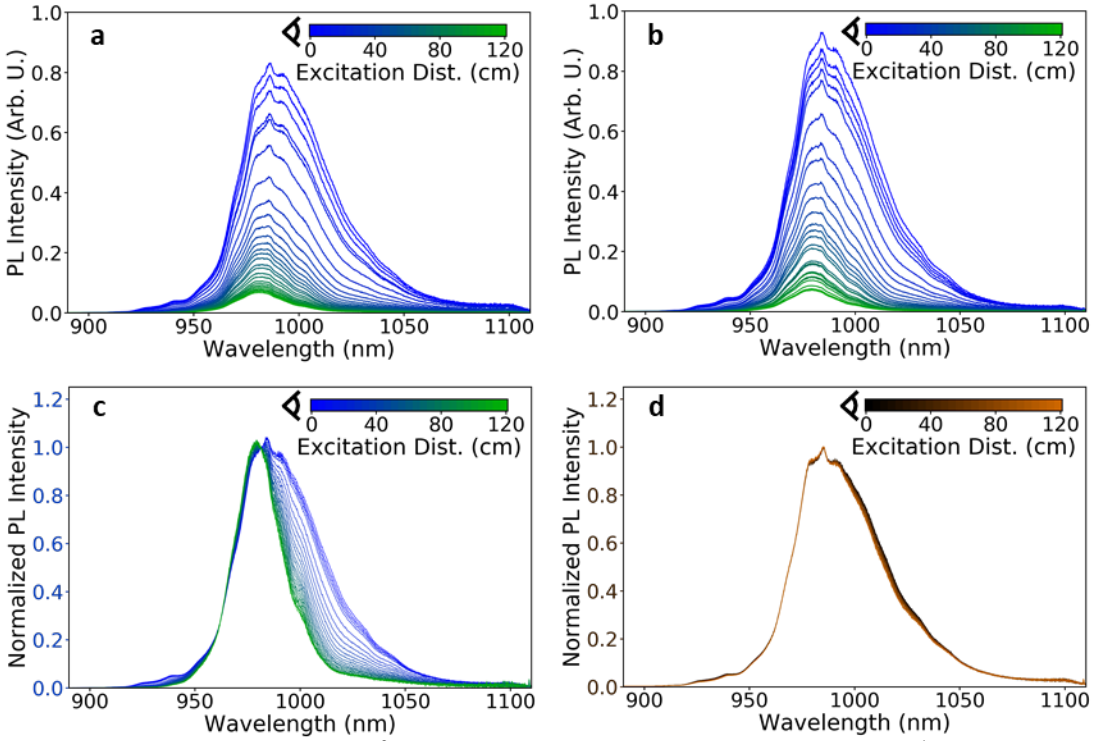

Figure S1. (a, b) PL spectra of Yb^{3+}:$CsPbCl_3$ NCs with **(a)** $OD_t \sim 0.75\ mm^{-1}$ at 375 nm and **(b)** $OD_t \sim 0.075\ mm^{-1}$ at 375 nm, suspended in hexane. Spectra were collected at various excitation distances in the 120 cm 1D LSC. **(c, d)** Normalized PL spectra of Yb^{3+}:$CsPbCl_3$ NCs with $OD_t \sim 0.075\ mm^{-1}$ at 375 nm suspended in **(c)** hexane and **(d)** TCE, collected at different excitation distances in the 1D LSC. The insets show the color coding with distance.

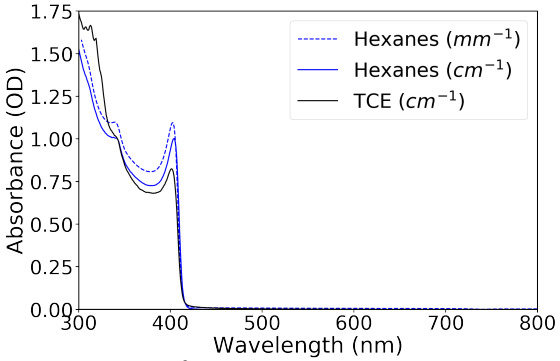

Figure S2. Absorption spectra of the Yb^{3+}:$CsPbCl_3$ nanocrystals used in the 1D LSC experiment of the main text. The blue dashed trace corresponds to the triangles reported in Figure 2b of the main text, the blue solid trace corresponds to the circles reported in Figure 2b of the main text, and the solid black trace corresponds to the NCs in TCE shown in Figure 3 of the main text.

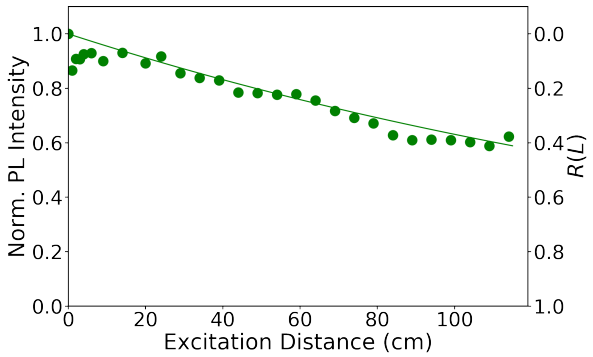

Figure S3. Waveguide attenuation data plotted as PL intensity *vs* excitation distance. The green curve shows the result of fitting the data using eq 1 of the main text, with a wavelength independent extinction coefficient of 0.002 cm^{-1}.

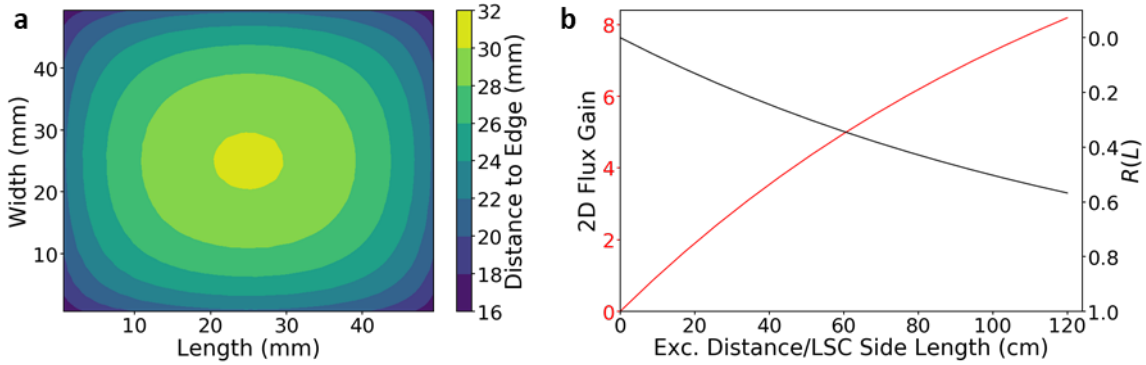

Figure S4. (a) Average distance to the edge of a 50 x 50 x 1 mm square from any point in the square. Averaging was performed over polar angles from 42° to 138° and over all azimuthal angles. **(b)** Reabsorption probability and 2D flux gain simulation of an LSC containing $Zn_{0.87}Cd_{0.11}Mn_{0.02}Se/ZnS$ NCs obtained using the same methods developed for the models presented in the main text. Both curves are in good agreement with our previous report.[2]

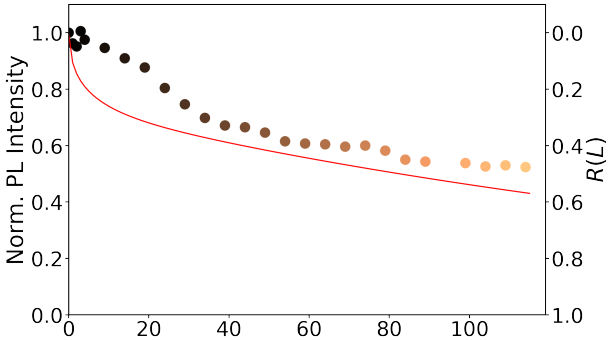

Figure S5. Integrated normalized PL intensity of Yb^{3+}:$CsPbCl_3$ NCs plotted as a function of excitation distance in the 1D LSC for solutions in TCE with an $OD_t \sim 0.075$ mm^{-1} (black-brown). The red trace shows the reabsorption probability as a function of excitation distance for the $CuInS_2/ZnS$ NCs used in modeling, obtained from the absorption and PL spectra shown in Figure 5b of the main text.

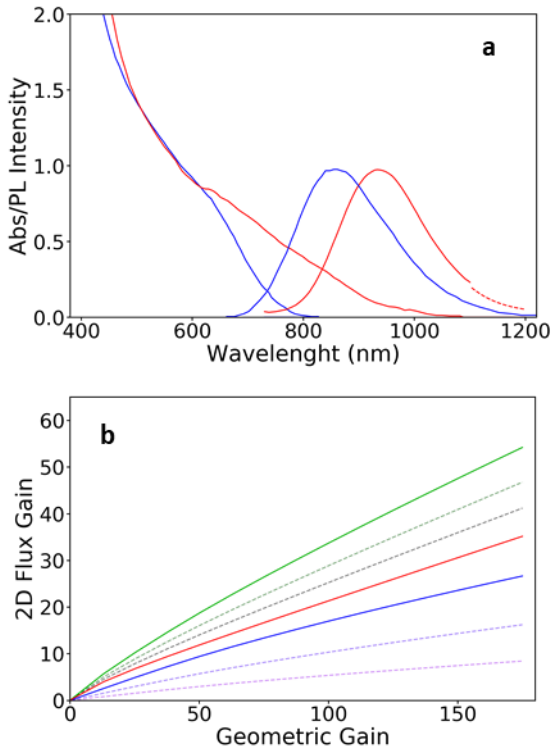

Figure S6. (a) Absorption and normalized PL spectra of high-quality $CuInS_2/ZnS$ NCs from literature[3] (blue traces) and of QD-950 NCs from the Strem catalog[4] (red traces). PL data above 1100 nm were not provided in ref [4], so the PL intensity below 1100 nm was extrapolated by plotting the PL on an energy x-axis and fitting the available PL data to a gaussian curve. Data extrapolated from the gaussian fit is shown as a dashed red curve. **(b)** Projected 2D flux gain of an Yb^{3+}:$CsPb(Cl_{1-x}Br_x)_3$ NC LSC (purple to blue gradient), a QD-950 NC LSC with a PLQY of 91% (red), and the monolithic bilayer device shown in Figure 5a of the main text (black to green gradient), using the QD-950 absorbance and PL intensity data shown in panel (a). The absorption onset of the Yb^{3+}:$CsPb(Cl_{1-x}Br_x)_3$ NCs is varied linearly from 412 to 488 nm for the three plotted traces.

References

1. R. Sumner, S. Eiselt, T. B. Kilburn, C. Erickson, B. Carlson, D. R. Gamelin, S. McDowall and D. L. Patrick, *J. Phys. Chem. C*, 2017, **121**, 3252-3260.
2. L. R. Bradshaw, K. E. Knowles, S. McDowall and D. R. Gamelin, *Nano Lett.*, 2015, **15**, 1315-1323.
3. M. R. Bergren, N. S. Makarov, K. Ramasamy, A. Jackson, R. Guglielmetti and H. McDaniel, *ACS Energy Lett.*, 2018, **3**, 520-525.
4. Copper Indium DiSulfide/Zinc Sulfide Quantum Dots, https://www.strem.com/ubiqd.