Supporting Information

FRET-Driven Hybrid Polymer–Perovskite Matrices for Efficient Pure-Red Emission

Jyoti Saxena,*[a] Rahul Murali,[b] Avari Roy,[a] Abdullah A. Al-Kahtani,[c] Venugopal Rao Soma,[d] Sai Santosh Kumar Raavi,[b] and Aditya Sadhanala*[a,c]

Corresponding author: Jyoti Saxena – jyotiphysics06@gmail.com

Aditya Sadhanala - sadhanala@iisc.ac.in

a Centre for Nano Science and Engineering, Indian Institute of Science, Bengaluru 560012, Karnataka, India

b Ultrafast Photophysics and Photonics Laboratory, Department of Physics, Indian Institute of Technology Hyderabad, Kandi 502285, Telangana, India

c King Saud University, College of Science, Chemistry Department, P. O. Box 2455, Riyadh-11451, Saudi Arabia

d School of Physics, University of Hyderabad, Hyderabad 500046 Telangana, India; DRDO Industry Academia - Centre of Excellence (DIA-CoE; formerly ACRHEM), University of Hyderabad, Hyderabad 500046 Telangana, India.

Experimental Details:

***Materials*.** Lead iodide (PbI2) (99.99%) was purchased from TCI Chemicals and methylammonium bromide (MABr) from great cell solar materials. Zinc acetate dihydrate, oleic acid (99.0%), oleylamine (70%), 2-methoxyethanol, ethanolamine, toluene, methyl acetate, chlorobenzene (CBZ), and chloroform (CLF) were purchased from Sigma-Aldrich. Methylamine (MA) dissolved in acetonitrile (ACN) solution was purchased from TCI Chemicals. CBP and TFB were purchased from Oscillas (Tokyo, Japan). All purchased chemicals were used as received.

***Synthesis of Lead Mixed Halides Red Emitting Perovskite Nanocrystals*.** For the synthesis of $MAPbI_2Br$ PeNCs, a perovskite precursor solution was prepared by dissolving 2 mmol PbI_2 and 2 mmol MABr in 4 mL MA dissolved in ACN solution[1]. Then, we synthesized the PeNCs using the ligand-assisted reprecipitation (LARP) technique by following the synthesis protocol described in,[1] with some modifications. Briefly, 0.2 mL a perovskite precursor solution was injected into a mixture of toluene (5 mL), oleic acid (1.8 mL), and oleylamine (0.2 mL), while stirring at 65°C. After injection, the reaction mixture was stirred for 90 s before cooling in an ice-water bath. The PeNCs were then purified three times by precipitation using an antisolvent methyl acetate at a ratio of 1:3 v/v. The supernatant was discarded, and the PeNCs were redispersed in toluene. The PeNCs were centrifuged at 5000 rpm for 5 min to remove aggregates and large particles before any further use of the PeNCs for characterizations.

Synthesis of CBP, TFB, and PeNCs ternary blend. The ternary blend was prepared by physically mixing CBP (10 mg/mL in equal volumes of CBZ and CLF) and TFB (10 mg/mL in equal volumes of CBZ and CLF) in a ratio 1:1. PeNCs (dispersed in toluene) were added at ratios of 1:3, 1:5, and 1:8 to this binary blend. All the above-mentioned ratios were fixed based on the most efficient energy transfer phenomenon observed in the photoluminescence studies performed on various ratios of the ternary blend.

TEM and HRTEM. The size distributions of the mixed lead halide perovskite NCs on a copper grid were determined by transmission electron microscopy (TEM) using a TITAN Themis TEM with a LaB6 filament operating at 300 kV for both low-magnification (TEM) and high-resolution (HRTEM) images.

***UV–vis Absorption*.** The absorption spectra of the solid-state films were recorded using a commercial UV-Vis NIR PerkinElmer LAMBDA 1050.

***Steady-State Photoluminescence (PL)*.** Steady-state PL measurements were carried out using a Ocean Insight QEPro High Performance spectrometer. The samples were excited by a diode laser of wavelength 405 nm. Energy transfer was characterized by photoluminescence spectra pumped at a wavelength of 405 nm.

***Time-Resolved Photoluminescence (TRPL)*.** TRPL measurements were performed using a commercial FLS1000 Photoluminescence Spectrometer. The setup was operated in time-correlated single-photon counting (TCSPC) mode. Pulsed laser diodes with a wavelength of 375 nm were used for excitation.

Morphology Characterization. Non-contact-mode atomic force microscopy images were obtained using a Park AFM NX20.

***Femtosecond Transient Absorption spectroscopy*.** Femtosecond transient absorption spectroscopy (fs-TAS) measurements were performed using a HELIOS pump-probe spectrometer (Ultrafast Systems). A detailed explanation of this setup can be found in literature[2,3]. In brief, a Ti:Sapphire amplifier (LIBRA, M/s Coherent, USA) was used as the laser source, giving 4mJ pulses with a repetition rate of 1kHz, central wavelength of 800 nm, and pulse duration of ~ 70 fs. The output pulses were split into two beams using a beam splitter: the intense beam (pump) passed through a BBO crystal to produce a 400 nm pump, whereas the low-intensity beam (probe) was focused onto a 3 mm thick sapphire window to produce a white-light continuum from 450 to-780 nm. A Helios optical chopper was used to reduce the pump frequency to 500 Hz, and a retroreflector mounted on a motor stage was used to control the optical delay of the probe. All films were encapsulated inside the glove box before performing the TAS measurements to ensure that there was no sample degradation. The encapsulated films were then placed at the focus of the pump and probe beams. The TAS measurements for all the samples were conducted at a pump intensity of $10\ \mu J/cm^2$. The obtained data were chirp-corrected and further analyzed using the Surface Xplorer software (Ultrafast Systems).

***Photothermal Deflection Spectroscopy (PDS)*.** Photothermal Deflection Spectroscopy (PDS) was performed using our custom-built setup. A 350W Xe lamp served as the pump beam source, while a 670 nm laser was used as the fixed-wavelength probe. The probe beam deflection was detected by a four-quadrant photodiode (Hamamatsu Photonics). In this technique, the sample is immersed in a temperature-sensitive photothermal liquid perfluoro hexane. When the pump beam heats the sample, it releases heat that creates a local temperature gradient in the liquid. This gradient deflects the grazing-incidence probe beam (like the mirage effect), and the deflection measured by the detector is directly proportional to the sample’s absorption.

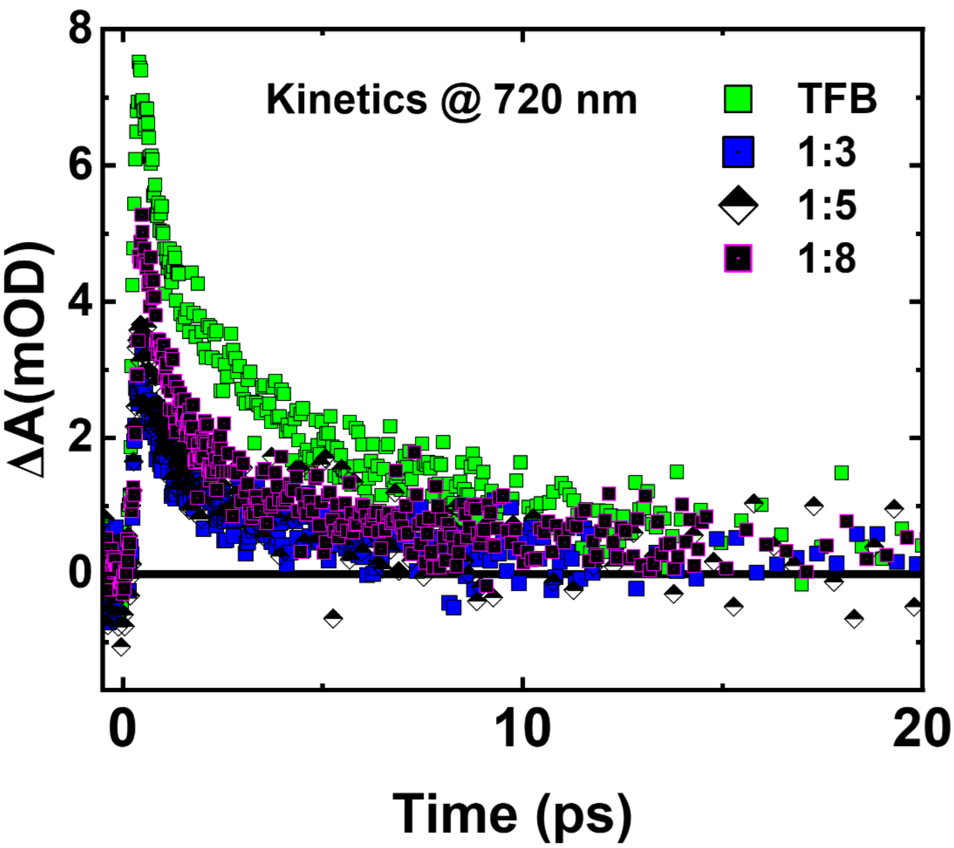

Figure S1: Kinetics at photoinduced absorption (PIA-2), scatter points denote the experimental data.

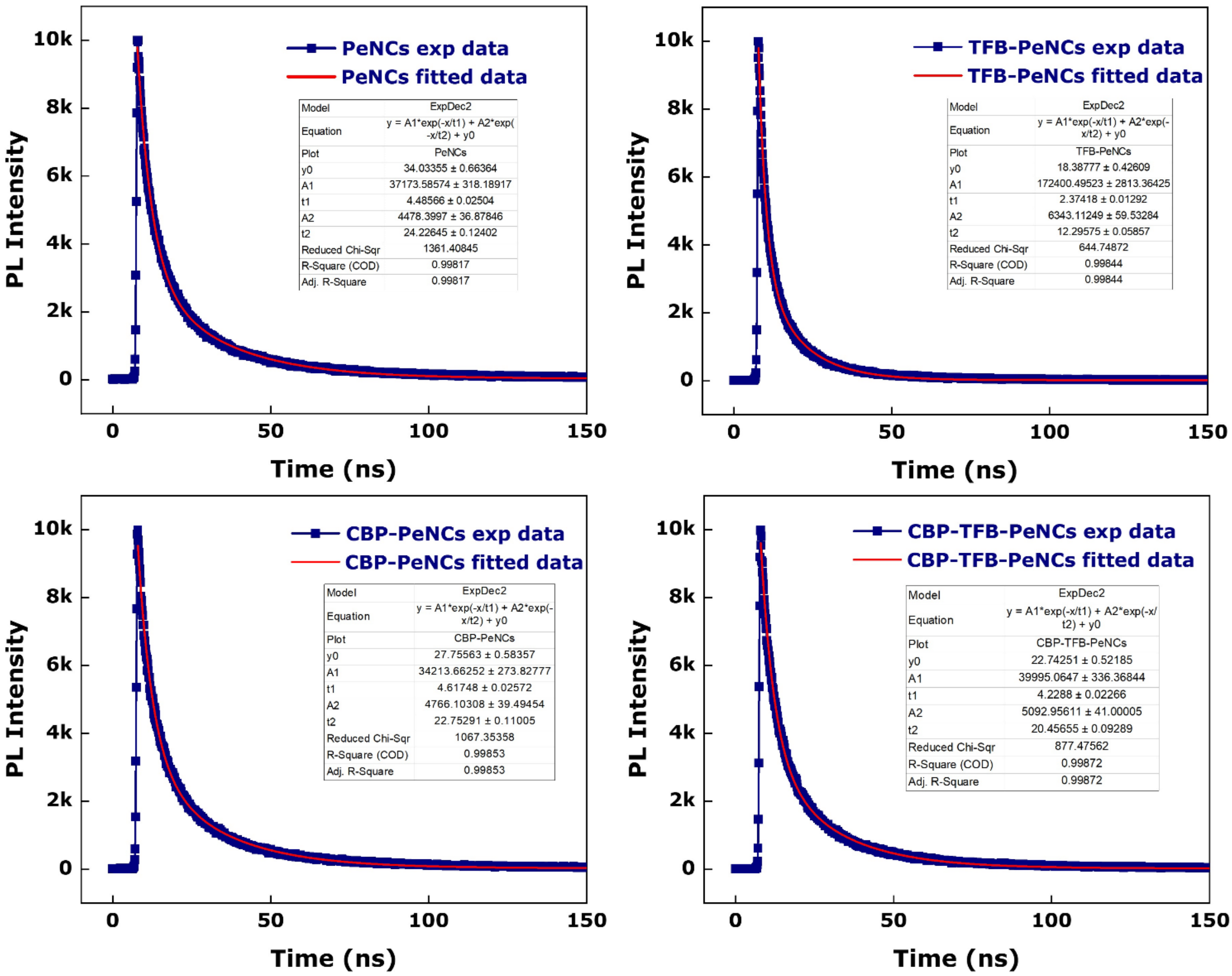

Figure S2: Lifetime measurements of PeNCs, PeNCs in TFB, PeNCs in CBP, PeNCs in CBP and TFB. Blue lines represent experimental data and the red lines represent fitted data.

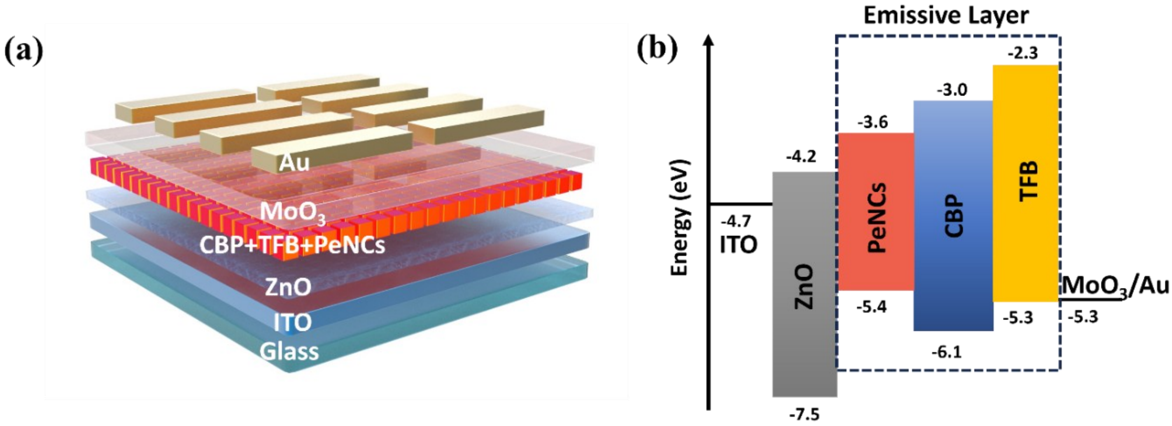

Figure S3. **(a)** Proposed device architecture for PeLEDs and **(b)** flat-band energy-level diagram of the proposed device architecture, the HOMO and LUMO energy levels of CBP and TFB are taken from T. Zhang et. al.[4].

References

1 Y. Hassan, O. J. Ashton, J. H. Park, G. Li, N. Sakai, B. Wenger, A. A. Haghighirad, N. K. Noel, M. H. Song, B. R. Lee, R. H. Friend and H. J. Snaith, *J Am Chem Soc*, 2019, **141**, 1269–1279.

2 R. Murali, C. Biswas, S. K. Nayak, H. Wu, X. Peng, V. Kumar, P. Chetti, V. R. Soma and S. S. K. Raavi, *J Mater Chem C Mater*, DOI:10.1039/D4TC03281K.

3 C. Biswas, S. G. Palivela, L. Giribabu, V. R. Soma and S. S. K. Raavi, *Opt Mater (Amst)*, 2022, **127**, 112232.

4 T. Zhang, M. Long, L. Pan, K. Ngai, M. Qin, F. Xie, X. Lu, J. Chen and J. Xu, *Sci Bull (Beijing)*, 2020, **65**, 1832–1839.