Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Unprecedented Random Lasing in Morphologically Tuned Organolead Halide Hybrid 2D Single Crystal Perovskite Microrods

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The slow cooling technique was initially used to grow the Organolead halide FA-(N-MPDA)$PbBr_4$ perovskite. The supersaturated solution of the FA-(N-MPDA)$PbBr_4$ was kept into the oil bath and the temperature was lowered at a constant rate of 1°C/hr to room temperature. By this slow cooling technique, we observed that the random crystallization with multi-crystal stacking was formed rapidly as soon as the solution reached supersaturation (**Figure S1a**). Evaporation of solvent from the solution creates a high local supersaturation and generates unwanted nuclei to form tiny crystals. These tiny crystals fall into the solution and hamper the growth of the crystals. Overall, the crystals grown by this method are small and the shapes are unpredictable. On the other hand, in slow evaporation at constant temperature (SECT) technique the nucleation was controlled systematically by maintaining a particular temperature (40 °C) with controllable slow evaporation of the supersaturated solution. In this case, the SECT synthesis process efficiently leads to reduce the multi-crystalline stacking of 2D perovskite in order to obtain well-shaped single crystals with millimeter scale as shown in **Figure S1b**. Furthermore, the low-temperature solution growth offers a variety of morphologies and polymorphic forms of the same crystals by different growth conditions. The morphology of the 2D perovskite was tuned by maintaining ambient temperature condition and kept the reaction over 24 hours with slow evaporation in order to grow the micro rod structure of FA-(N-MPDA)$PbBr_4$ perovskite (**Figure S1c**).

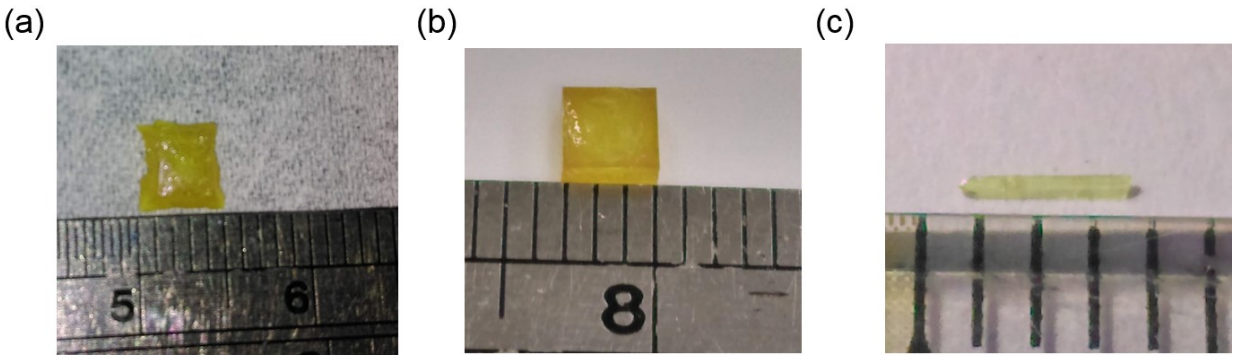

Figure S1. Organolead halide perovskite of FA-(N-MPDA)$PbBr_4$ crystals grown by using (a) slow cooling technique and (b & c) SETC technique.

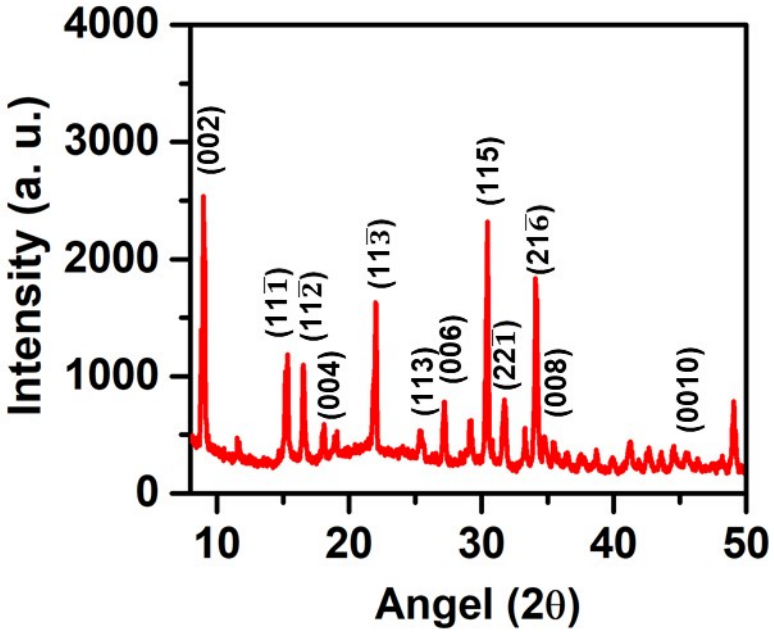

Figure S2. Standard XRD pattern of FA-(N-MPDA)$PbBr_4$

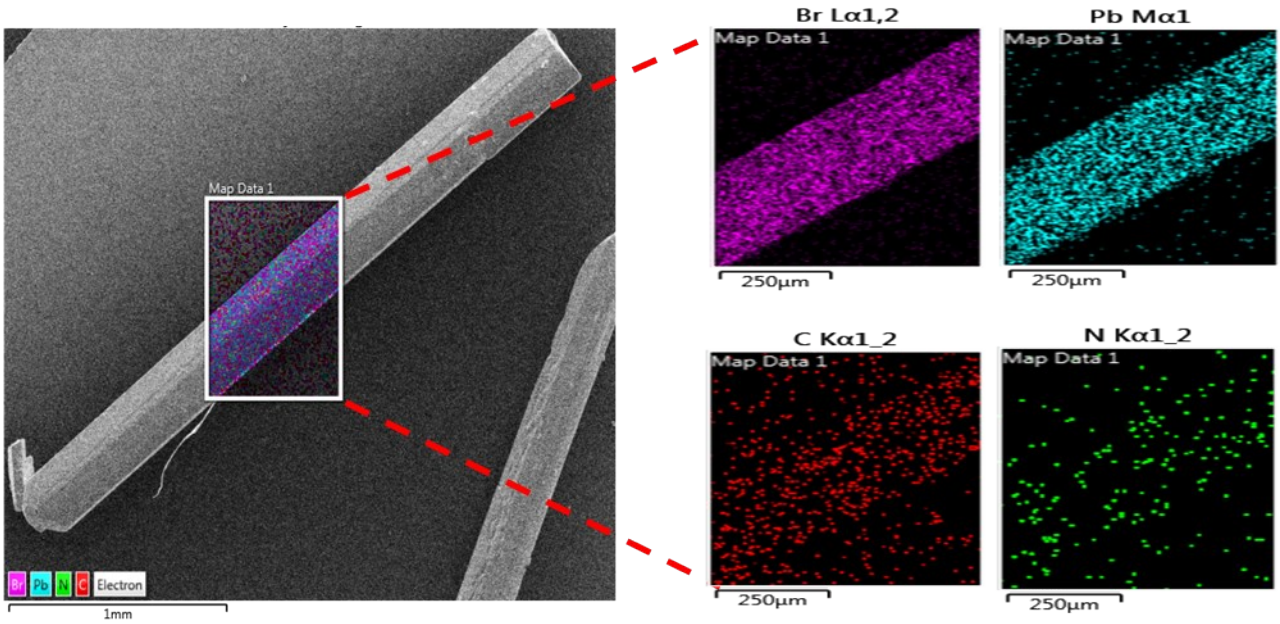

Figure S3. EDX mapping of FA-(N-MPDA)$PbBr_4$ microrod with the corresponding composition.

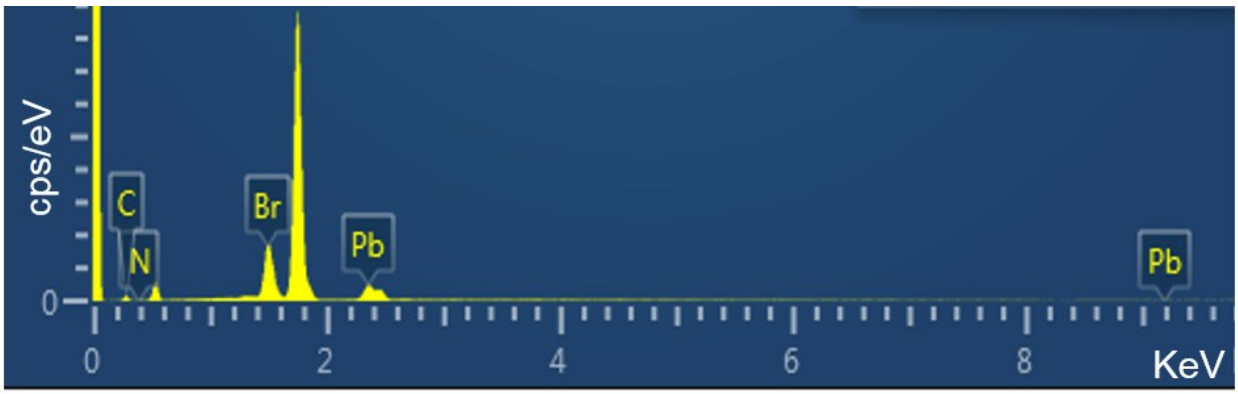

Figure S4. EDX spectrum of FA-(N-MPDA)$PbBr_4$ single crystal microrod with composition .

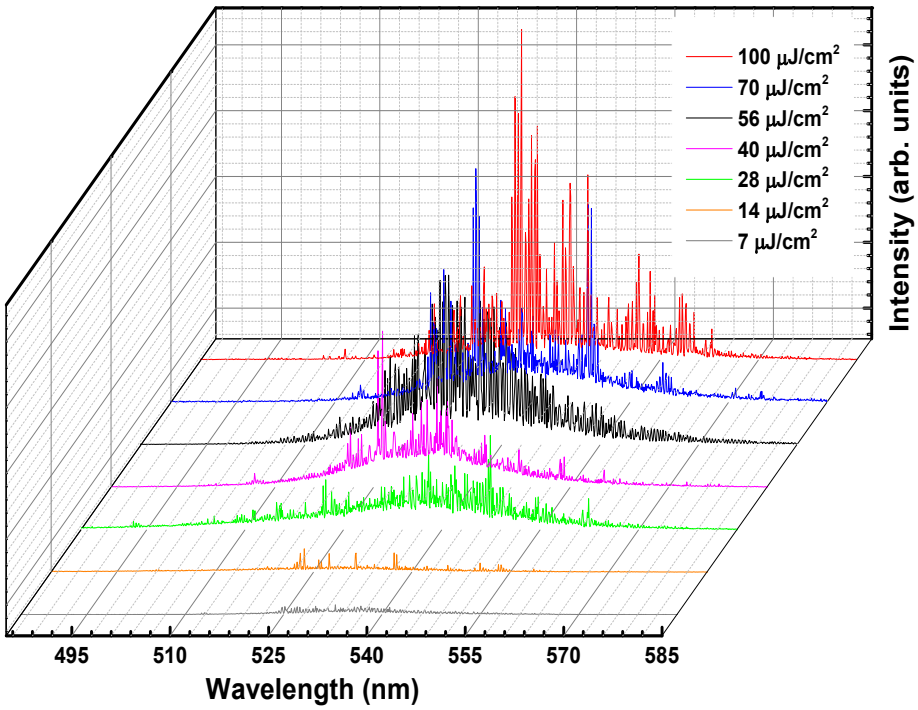

Figure S5. Random lasing from 2D perovskite single crystal microrod. Evolution of the emission spectra of 2D perovskite device as a function of pumping energy density.

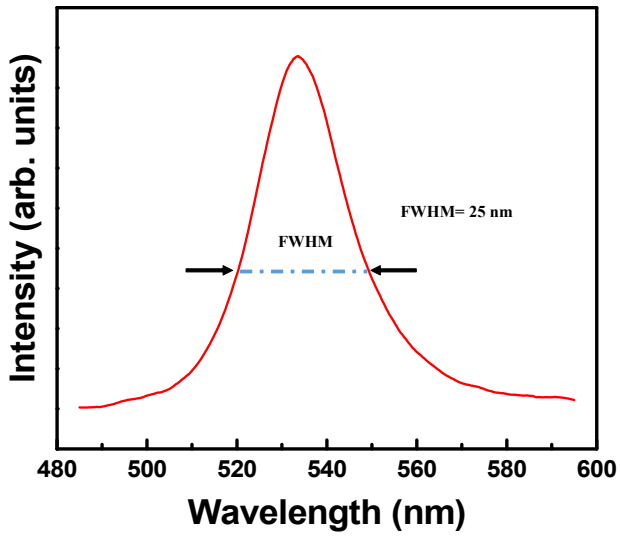

Figure S6. The PL emission spectrum of single-crystal 2D perovskite with a pump−fluence less than the threshold power density.

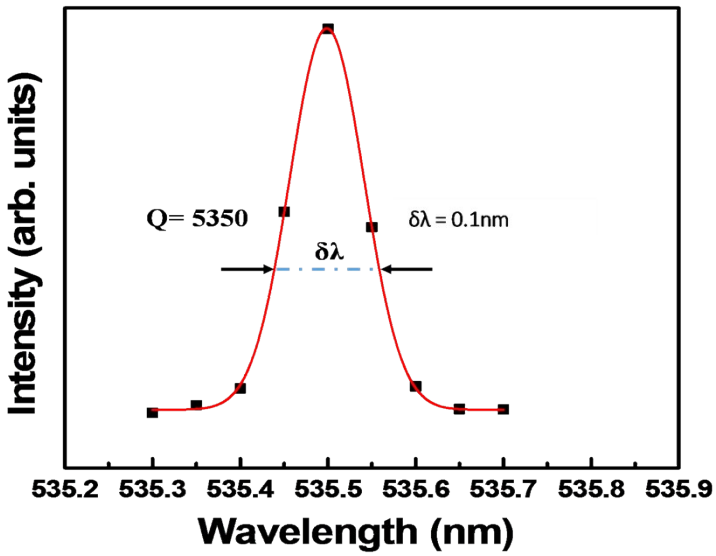

Figure S7. The Lorentzian fitting of a lasing oscillation mode. The FWHM of the lasing peak (δλ) is 0.1 nm, with a Q factor of ~5350.

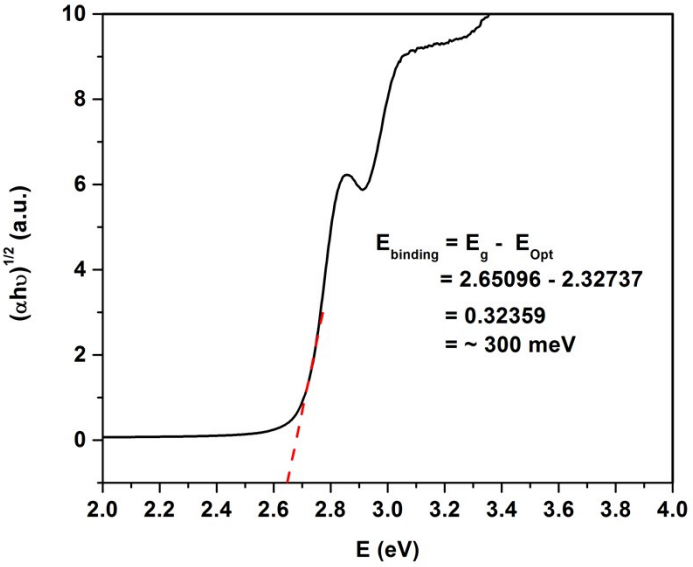

Figure S8. The tauc plot to determine the optical bandgap. Overlaid text shows the calculation of the binding energy of excitons in our system.

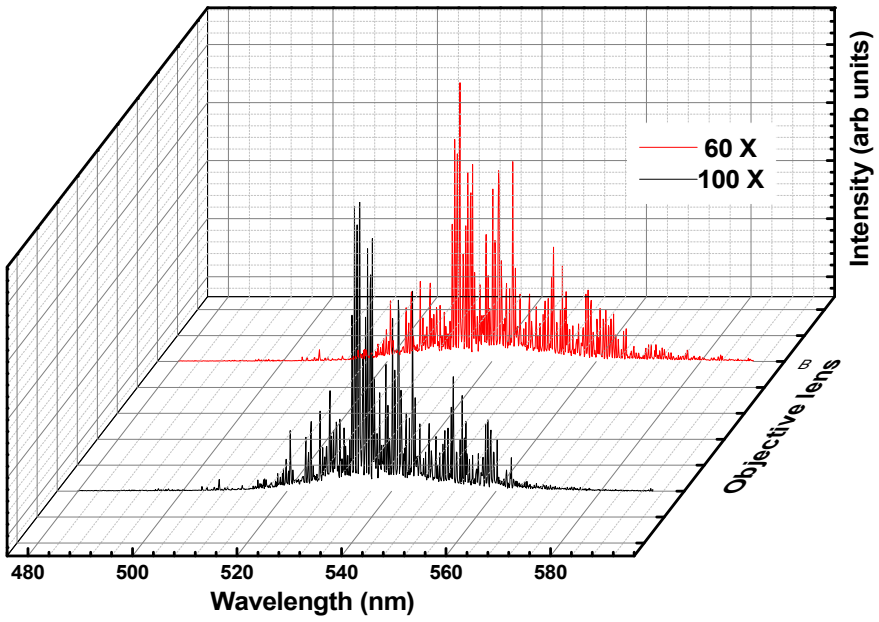

Figure S9. Variation of the laser spot size of the emitted spectrum while keeping the power density fixed at 12 μJ/ cm^2

Q factor Calculation:

Quality factor (Q) = λ/δλ,

where λ is the wavelength of the lasing peak

δλ is the line width of the fitted Gaussian curve during lasing

In our case, λ= 535 nm δλ= 0.10 nm

Therefore Q = 535/0.10

= 5350