Supporting Information

Dynamics of antisolvent processed hybrid metal halide perovskites studied by *in situ* photoluminescence and its influence on optoelectronic properties

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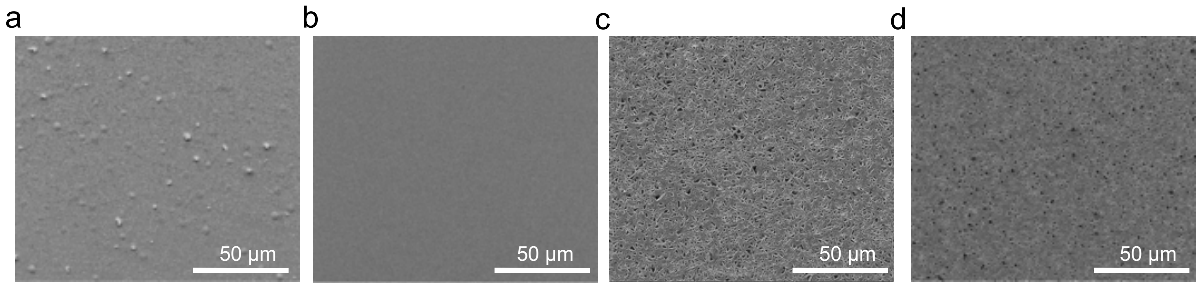

Figure S1. SEM images of $MAPbI_3$ films prepared by dripping the antisolvent after a) 3 s, b) 5 s, c) 7 s after the second spin cycle has started, and d) without antisolvent, respectively. Total annealing of 10 min at 100 ºC.

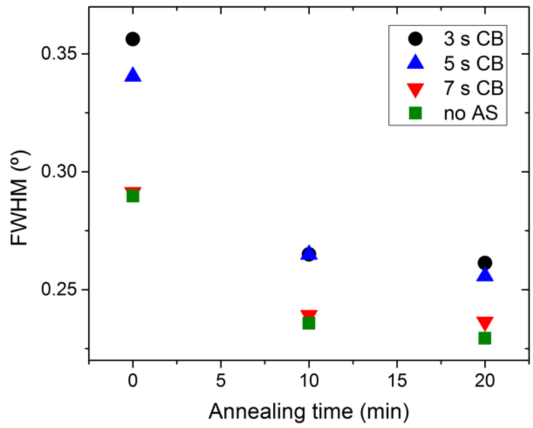

Figure S2. Full width at half maximum (FWHM) of the (110) $MAPbI_3$ diffraction peak after dripping the antisolvent at the time specified, and without application of an antisolvent (total of 0-20 min annealing at 100 ºC).

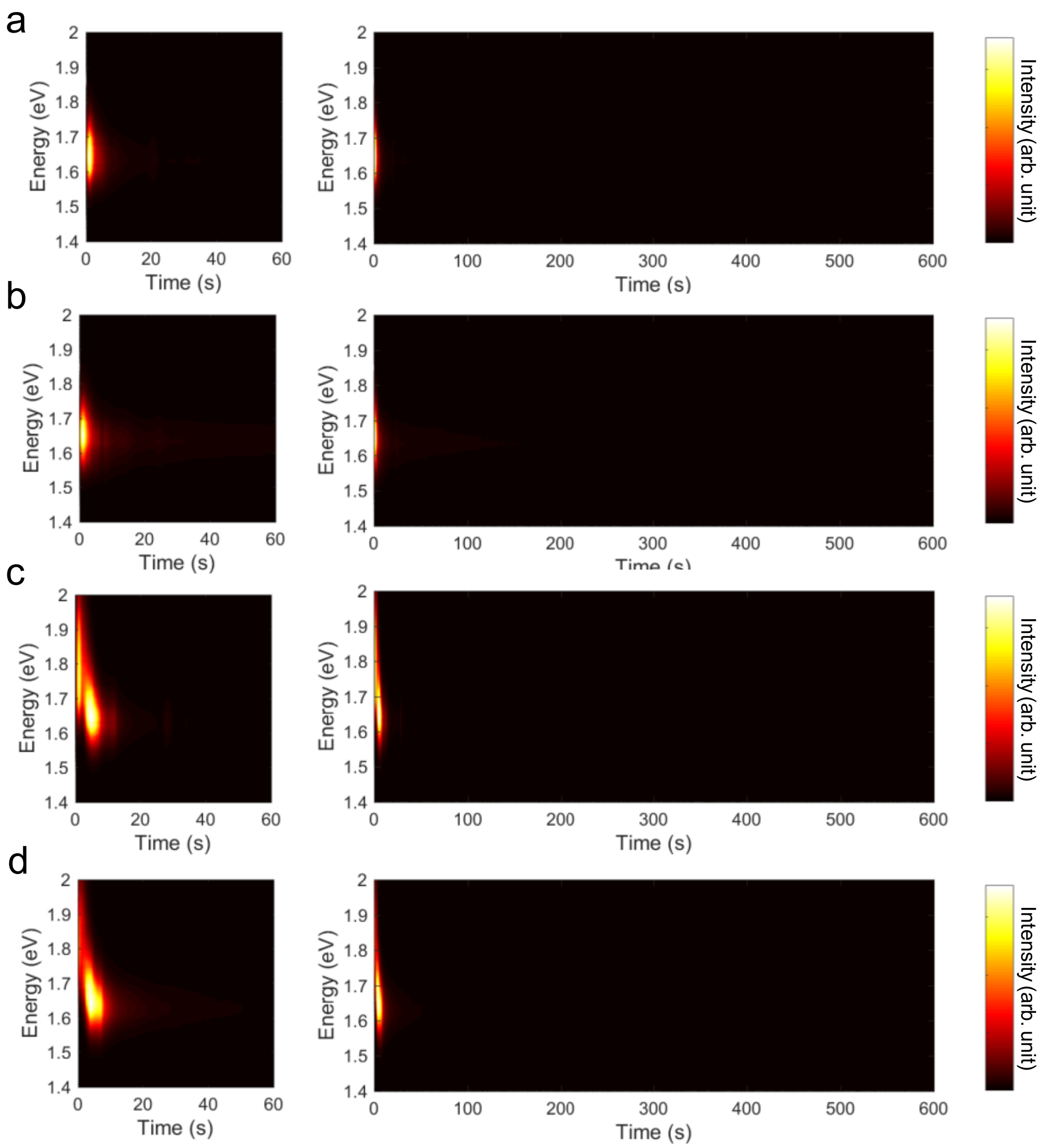

Figure S3. *In situ* PL contour plot of $MAPbI_3$ films taken on the hot plate (100 ºC) after dripping the antisolvent after a) 3 s, b) 5 s, c) 7 s and d) without antisolvent dripping.

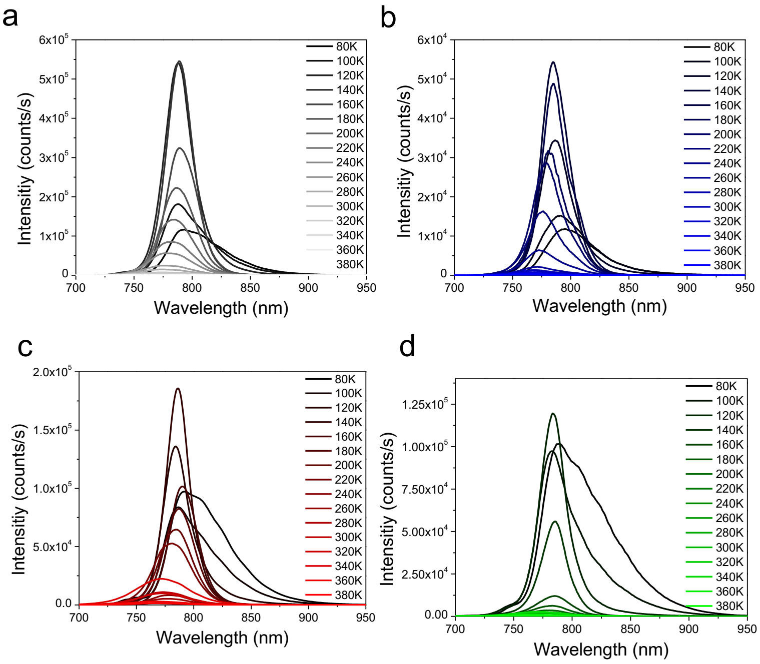

Figure S4. Low temperature PL measurements taken on samples with a) 3 s, b) 5 s, and c) 7 s antisolvent drip time and d) a reference sample without antisolvent dripping.

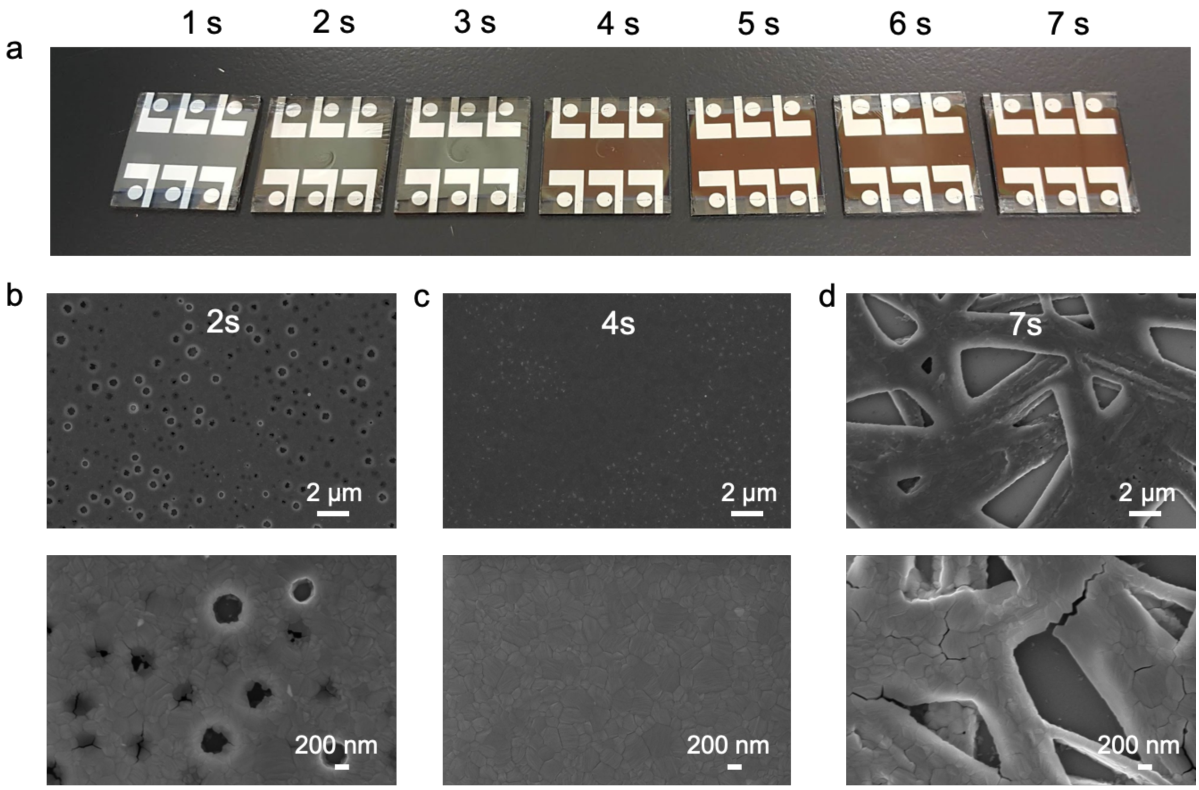

Figure S5. a) Picture of the $MAPbI_3$ films on ITO/NiO_x fabricated with different antisolvent drip times as indicated on the top. **b-d)** Top-view SEM images of the devices using drip times of 2, 4, and 7s.

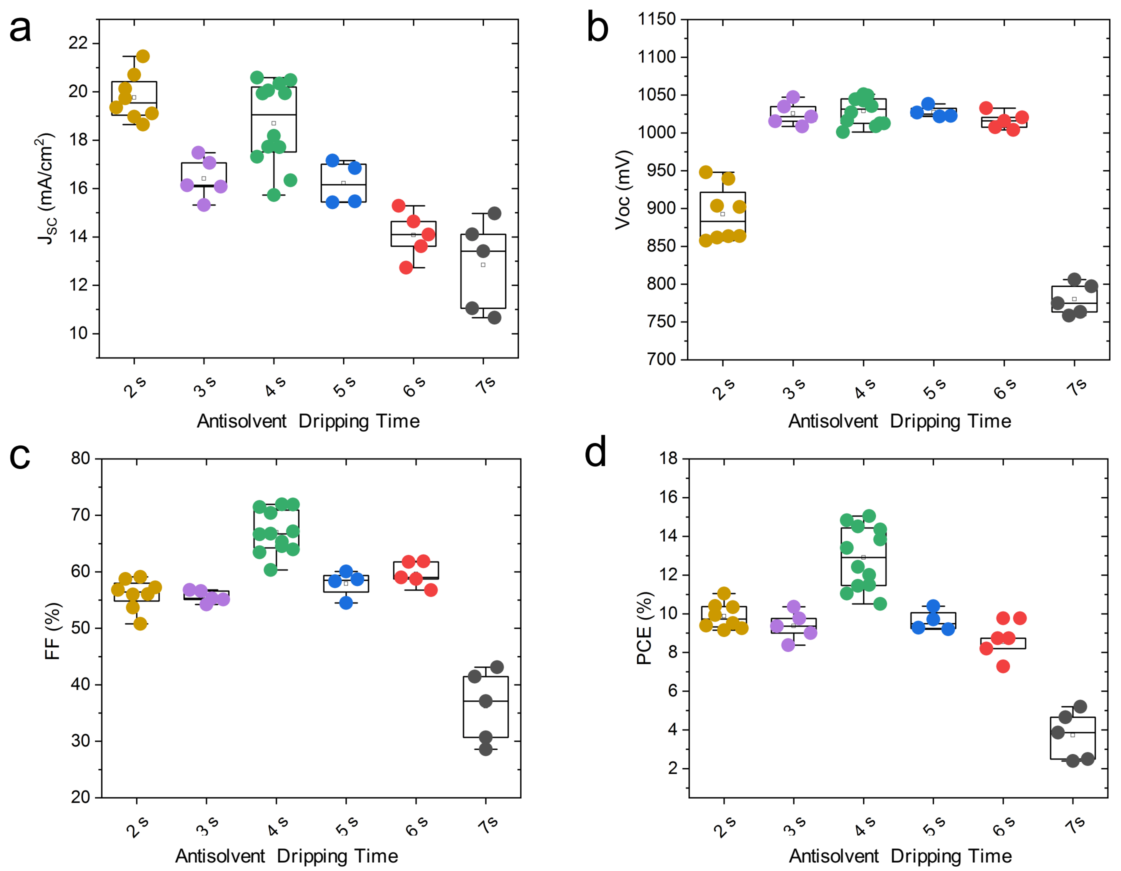

Figure S6. a-d) Summary of photovoltaic parameters using different antisolvent drip times. Open squares inside of the boxes indicate the mean values. Two independent batches confirm the very narrow distribution of parameters with good statistics and thus allow to extract the ideal dripping window at 4s.

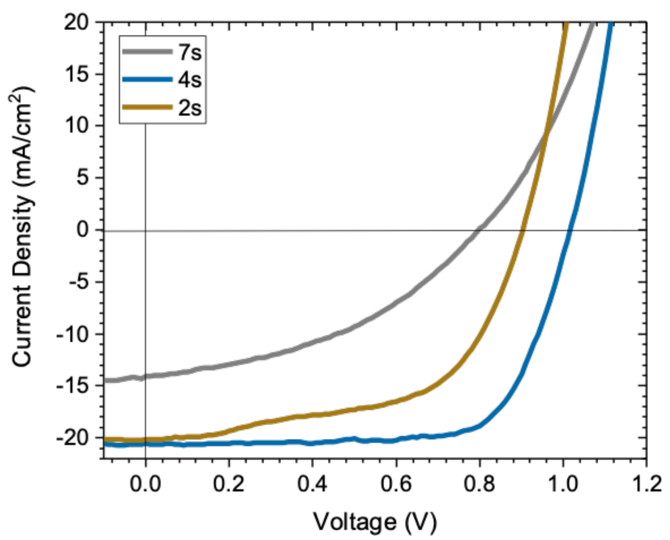

Figure S7. Representative J-V characteristics of devices using antisolvent drip times of 2, 4, and 7s.

Experimental Section

Experimental description of absolute PL quantum yields.

Absolute PL quantum yields were determined optically using a home-built integrating sphere spectrofluorometer. A Fianium SC450 supercontinuum pulsed laser is used as a white-light source with an average illumination intensity of 4 W from 410 nm to 2500 nm. The desired excitation wavelength is selected using two excitation monochromators, a Princeton Instruments SP150 and a Princeton Instruments SP275. Following wavelength selection, a small part of the excitation is directed to a ThorLabs S120VC calibrated silicon photodiode to measure the power, while the remainder is directed to a Spectralon integrating sphere from LabSphere, where it strikes the sample. The planar thin-film samples were oriented to avoid direct reflections back out of the entry port and prevent totally internally reflected light from leaving directly through the exit port, as these would yield incorrect PLQY values. A bare glass slide with the same orientation served as a blank. In addition to the tunable source, a second single wavelength laser was positioned to bypass the monochromators and enter through the same port of the integrating sphere. The second laser was a Power Technology LDCU 12/6692 laser that emitted at 440 nm with a max power of 16 mW. The second laser was also passed through a beamsplitter to a photodiode to monitor the power. PLQY values used in the paper used an excitation power of 9.7 mW over ~1 mm^2 and were calculated by integrating the counts of the $MAPbI_3$ thin film emission then comparing that value to the absorbed light at 440 nm. Direct reflections were blocked from exiting the sphere by a baffle, ensuring only diffuse light escaped the integrating sphere. The light exiting the sphere was focused onto the entrance slit of a Princeton Instruments SP2300 monochromator with a 300 g/mm grating blazed at 500 nm. The resultant spectrum was detected with a Princeton Instruments PIXIS 400 B thermoelectrically cooled silicon CCD camera. The CCD had been corrected for both spectral position using a Ne lamp and for sensitivity using a NIST-traceable radiometric calibration lamp from Ocean Optics, model HL3-plus, serial number 089440003. A complete description of this home-built integrating sphere spectrofluorometer can be found elsewhere.[1]

References

(1) Bronstein, N. D.; Yao, Y.; Xu, L.; O'Brien, E.; Powers, A. S.; Ferry, V. E.; Alivisatos, A. P.; Nuzzo, R. G. Quantum Dot Luminescent Concentrator Cavity Exhibiting 30-Fold Concentration. *ACS Photonics* **2015**, *2*, 1576–1583.