

## Supporting Information

# Effect of Density of Surface Defects on Photoluminescence Properties in MAPbI<sub>3</sub> Perovskite Films

*Paul Fassl<sup>a,b</sup>, Yuriy Zakharko<sup>c †</sup>, Lukas M. Falk<sup>a,b</sup>, Katelyn P. Goetz<sup>a,b</sup>, Fabian Paulus,<sup>a,b,c</sup>  
Alexander D. Taylor<sup>a,b</sup>, Jana Zaumseil<sup>b,c</sup> and Yana Vaynzof<sup>a,b \*</sup>*

<sup>a</sup> Kirchhoff Institute for Physics, Heidelberg University, 69120 Heidelberg, Germany

<sup>b</sup> Centre for Advanced Materials, Heidelberg University, 69120 Heidelberg, Germany

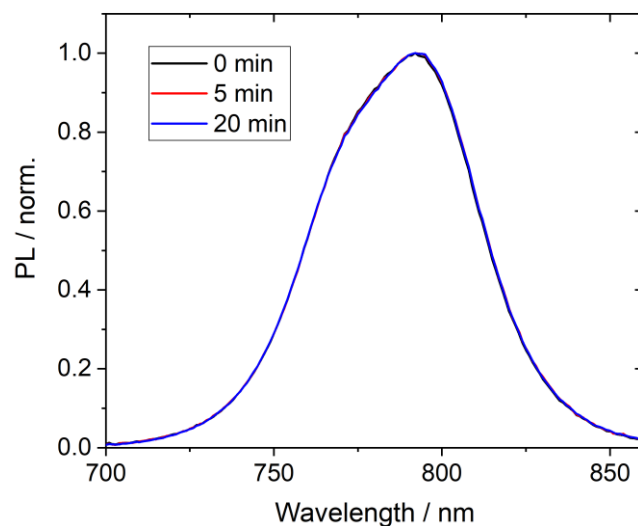
<sup>c</sup> Institute for Physical Chemistry, Heidelberg University, 69120 Heidelberg, Germany

<sup>†</sup> Present address: Department of Physics, Technical University of Denmark, 2800 Kongens

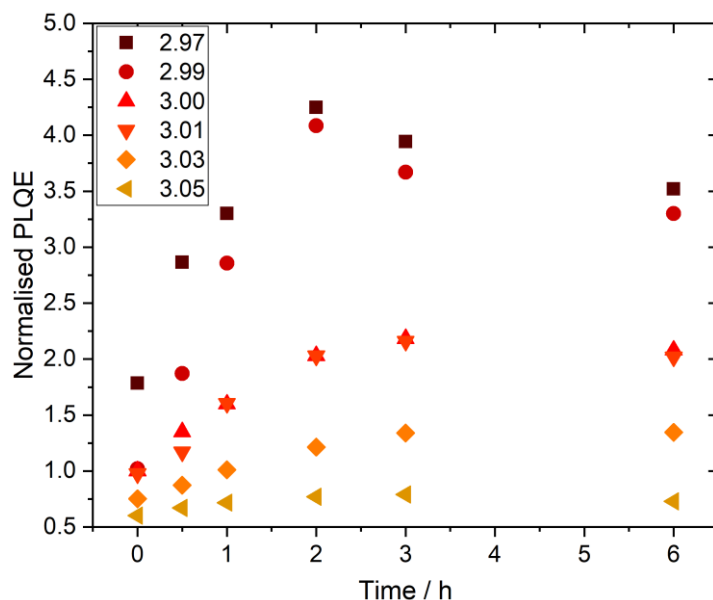
Lyngby, Denmark

### Corresponding Author

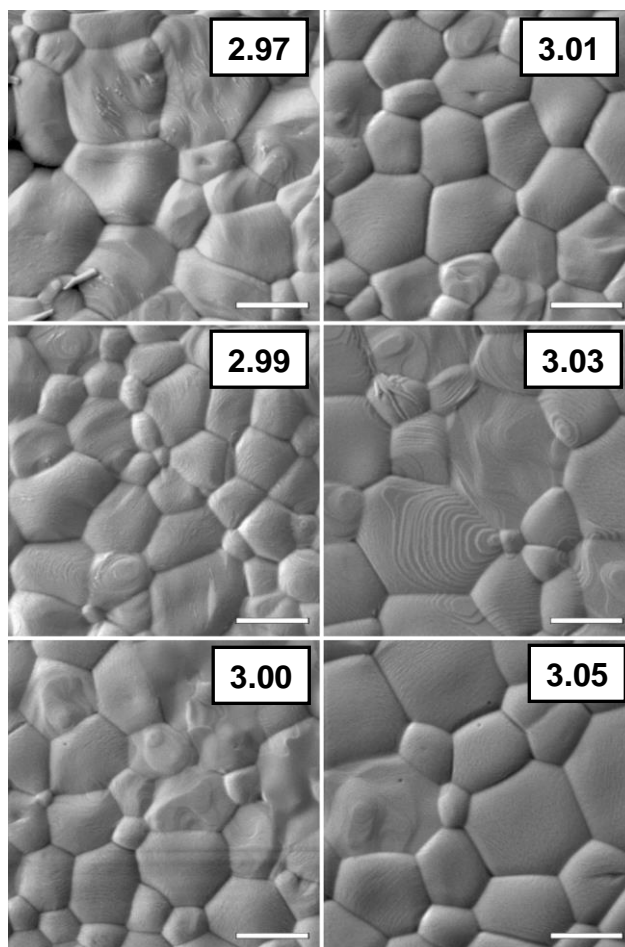
\* Email: [vaynzof@uni-heidelberg.de](mailto:vaynzof@uni-heidelberg.de).



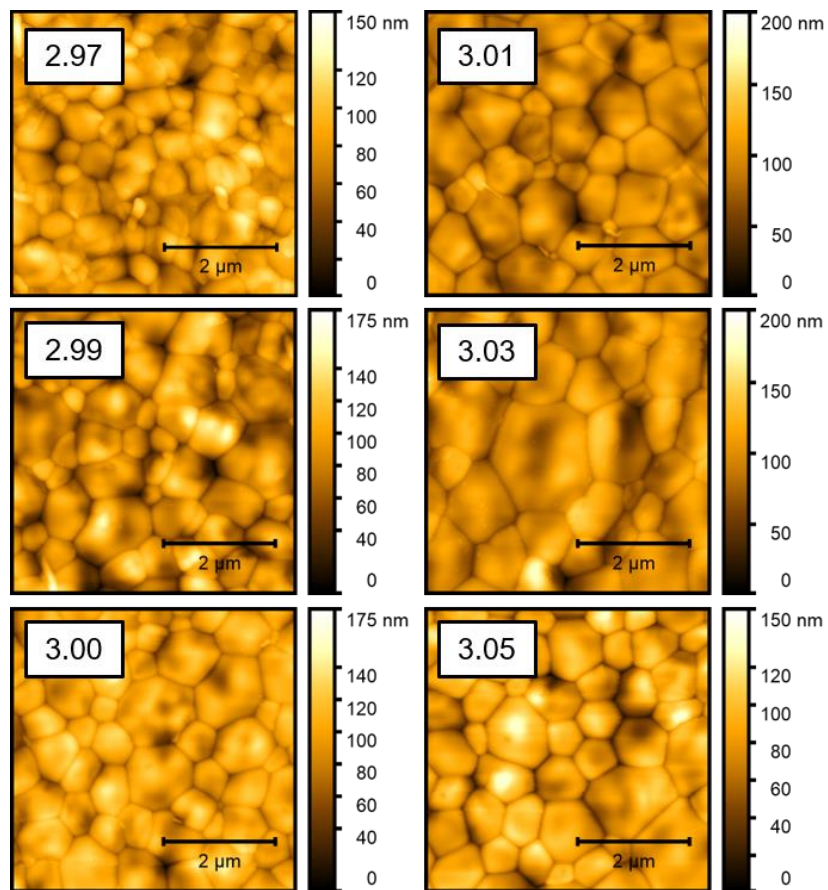
**Figure S1:** PL spectra of a glass/MAPbI<sub>3</sub> film with  $y = 3.03$  after 0, 5 and 20 minutes of illumination with a 532 nm CW laser with an intensity of 120 mW/cm<sup>2</sup>, measured inside an integrating sphere.



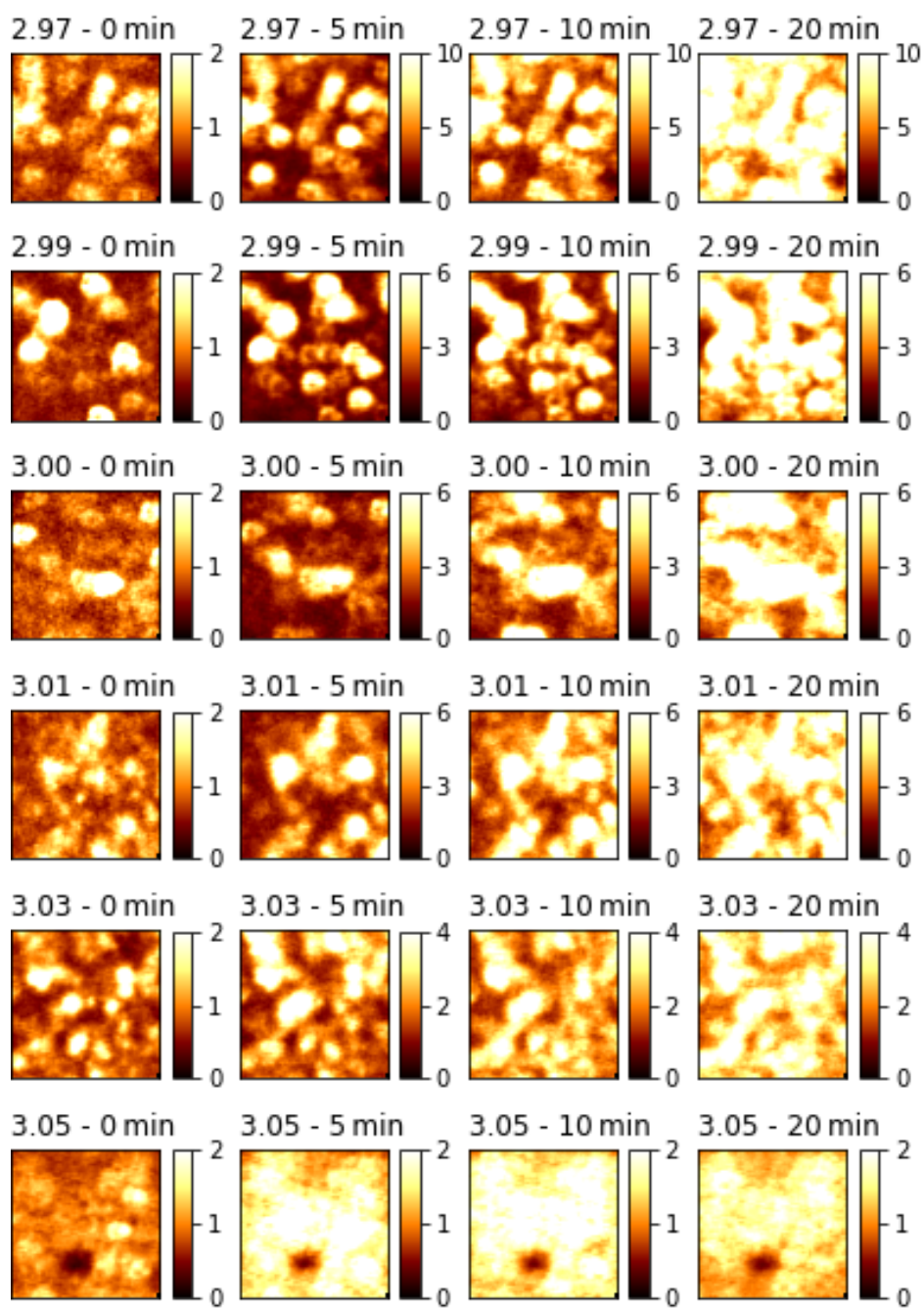
**Figure S2:** Temporal evolution of the PLQE upon long-term storage in ambient atmosphere and low levels of white light illumination for perovskite films fabricated from different stoichiometries  $y$ . The PLQE was normalized to the initial PLQE of the stoichiometric sample ( $y = 3.00$ ) for ease of comparison to other batches.



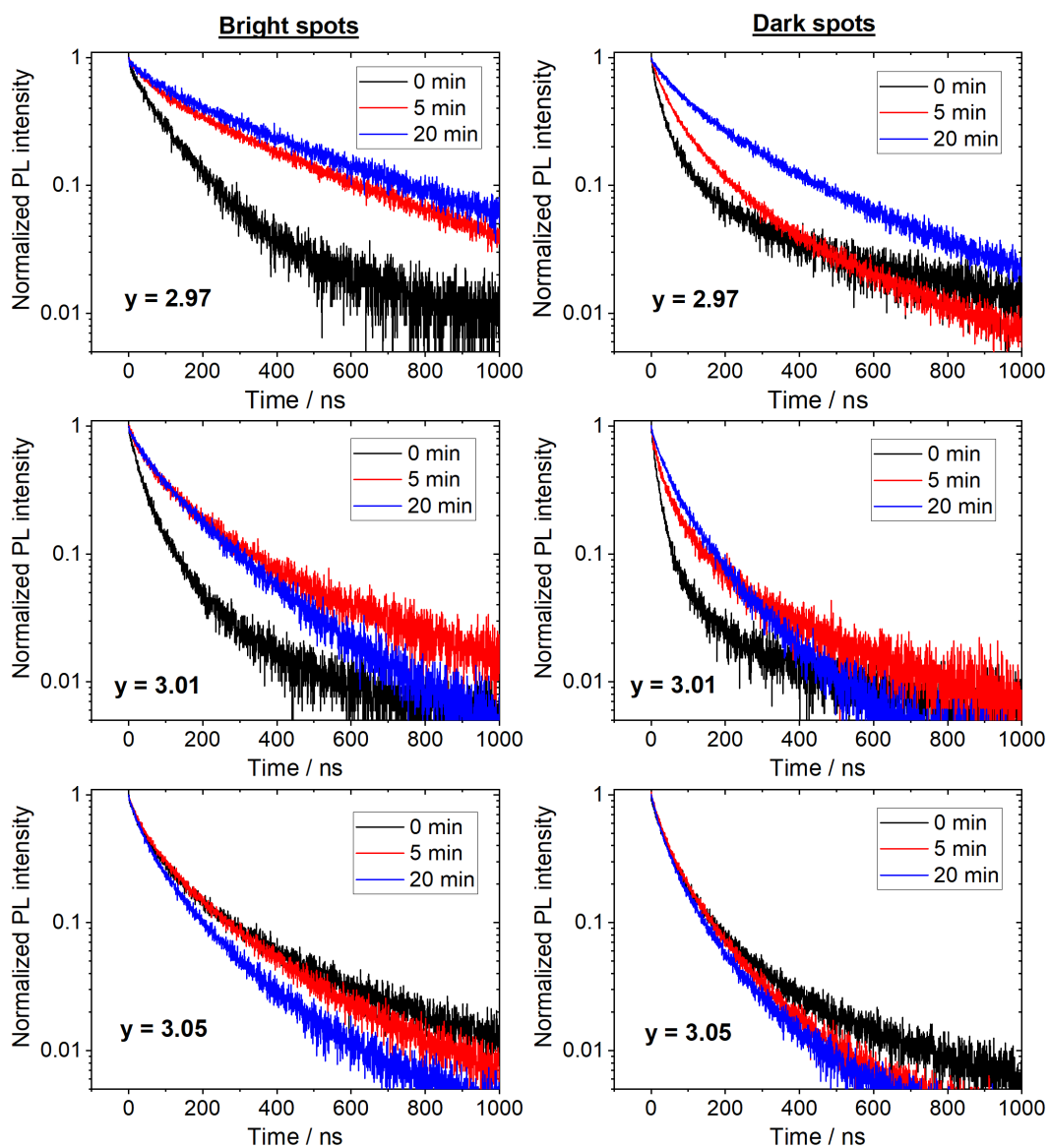
**Figure S3:** SEM images of perovskite films fabricated from different stoichiometries  $y$ . Scale bar is 1  $\mu\text{m}$ .



**Figure S4:** AFM micrographs of perovskite films fabricated from different stoichiometries  $y$ .



**Figure S5:** PL maps for all stoichiometries after 0, 5, 10 and 20 min of storage in ambient atmosphere and low levels of illumination, normalized to the average value of the 0 min map for each stoichiometry.



**Figure S6:** Representative PL decay data for stoichiometries  $y = 2.97/3.01/3.05$  after 0, 5 and 20 minutes for bright (left) and dark (right) regions (see Figure 3 in the main text).

**Table S1:** Fit results using equation (2) for the **bright** PL regions (see Figure 4).

Fit parameters \ Stoichiometry $y$		2.97	2.99	3.00	3.01	3.03	3.05
		0 min	$A_1$ (rel.)	0.02	0.08	0.10	0.44
$\tau_1$ [ns]	3		10	21	34	40	46
$A_2$ (rel.)	0.98		0.92	0.90	0.56	0.88	0.70
$\tau_2$ [ns]	52		79	51	44	100	92
b	0.73		0.58	0.44	0.61	0.65	0.63
$\tau_{Av}$ [ns]	<b>89</b>		<b>114</b>	<b>122</b>	<b>52</b>	<b>125</b>	<b>106</b>
5 min	$A_1$ (rel.)	0.21	0.31	0.24	0.15	0.07	0.18
	$\tau_1$ [ns]	45	27	50	74	27	49
	$A_2$ (rel.)	0.79	0.69	0.76	0.85	0.93	0.82
	$\tau_2$ [ns]	247	209	100	91	207	89
	b	0.79	0.76	0.58	0.63	0.68	0.68
	$\tau_{Av}$ [ns]	<b>233</b>	<b>179</b>	<b>131</b>	<b>122</b>	<b>251</b>	<b>103</b>
10 min	$A_1$ (rel.)	0.12	0.25	0.1	0.18	0.07	0.22
	$\tau_1$ [ns]	37	32	33	87	30	40
	$A_2$ (rel.)	0.88	0.75	0.9	0.82	0.93	0.78
	$\tau_2$ [ns]	312	178	123	125	208	95
	b	0.79	0.73	0.63	0.69	0.72	0.74
	$\tau_{Av}$ [ns]	<b>312</b>	<b>171</b>	<b>161</b>	<b>147</b>	<b>241</b>	<b>97</b>
20 min	$A_1$ (rel.)	0.04	0.22	0.19	0.16	0.14	0.28
	$\tau_1$ [ns]	2	35	38	51	58	41
	$A_2$ (rel.)	0.96	0.78	0.81	0.84	0.86	0.72
	$\tau_2$ [ns]	245	174	186	116	196	77
	b	0.74	0.75	0.71	0.81	0.73	0.72
	$\tau_{Av}$ [ns]	<b>285</b>	<b>169</b>	<b>195</b>	<b>118</b>	<b>213</b>	<b>80</b>

**Table S2:** Fit results using equation (2) for the **medium** PL regions (see Figure 4).

Fit parameters \ Stoichiometry $y$		2.97	2.99	3.00	3.01	3.03	3.05
		<b>0 min</b>	$A_1$ (rel.)	0.32	0.34	0.19	0.49
	$\tau_1$ [ns]	43	34	36	26	39	48
	$A_2$ (rel.)	0.68	0.66	0.81	0.51	0.59	0.71
	$\tau_2$ [ns]	45	35	25	23	68	73
	b	0.54	0.48	0.43	0.53	0.66	0.65
	$\tau_{Av}$ [ns]	<b>68</b>	<b>60</b>	<b>61</b>	<b>34</b>	<b>69</b>	<b>85</b>
<b>5 min</b>	$A_1$ (rel.)	0.23	0.25	0.17	0.24	0.10	0.25
	$\tau_1$ [ns]	55	23	43	50	82	40
	$A_2$ (rel.)	0.77	0.75	0.83	0.76	0.90	0.75
	$\tau_2$ [ns]	125	144	58	70	115	90
	b	0.65	0.74	0.57	0.57	0.63	0.75
	$\tau_{Av}$ [ns]	<b>145</b>	<b>135</b>	<b>86</b>	<b>96</b>	<b>155</b>	<b>90</b>
<b>10 min</b>	$A_1$ (rel.)	0.10	0.25	0.15	0.19	0.15	0.28
	$\tau_1$ [ns]	48	29	47	40	86	39
	$A_2$ (rel.)	0.90	0.75	0.85	0.81	0.85	0.72
	$\tau_2$ [ns]	161	160	81	101	129	89
	b	0.64	0.76	0.61	0.69	0.66	0.77
	$\tau_{Av}$ [ns]	<b>207</b>	<b>149</b>	<b>110</b>	<b>113</b>	<b>160</b>	<b>86</b>
<b>20 min</b>	$A_1$ (rel.)	0.15	0.27	0.18	0.15	0.12	0.33
	$\tau_1$ [ns]	91	36	51	38	61	37
	$A_2$ (rel.)	0.85	0.73	0.82	0.85	0.88	0.67
	$\tau_2$ [ns]	229	168	121	94	120	80
	b	0.75	0.77	0.68	0.78	0.67	0.77
	$\tau_{Av}$ [ns]	<b>247</b>	<b>154</b>	<b>139</b>	<b>98</b>	<b>146</b>	<b>75</b>



**Table S3:** Fit results using equation (2) for the **dark** PL regions (see Figure 4).

Fit parameters \ Stoichiometry $y$		2.97	2.99	3.00	3.01	3.03	3.05
		<b>0 min</b>	$A_1$ (rel.)	0.45	0.30	0.24	0.66
	$\tau_1$ [ns]	26	17	13	17	28	41
	$A_2$ (rel.)	0.55	0.70	0.76	0.34	0.45	0.60
	$\tau_2$ [ns]	38	28	29	24	42	60
	b	0.47	0.42	0.43	0.48	0.55	0.62
	$\tau_{Av}$ [ns]	<b>57</b>	<b>62</b>	<b>63</b>	<b>27</b>	<b>45</b>	<b>69</b>
<b>5 min</b>	$A_1$ (rel.)	0.34	0.27	0.22	0.32	0.20	0.33
	$\tau_1$ [ns]	46	29	28	26	55	33
	$A_2$ (rel.)	0.66	0.73	0.78	0.68	0.80	0.67
	$\tau_2$ [ns]	80	44	37	41	63	65
	b	0.65	0.55	0.48	0.54	0.55	0.74
	$\tau_{Av}$ [ns]	<b>87</b>	<b>64</b>	<b>67</b>	<b>57</b>	<b>96</b>	<b>64</b>
<b>10 min</b>	$A_1$ (rel.)	0.14	0.22	0.18	0.31	0.14	0.35
	$\tau_1$ [ns]	66	37	36	33	47	31
	$A_2$ (rel.)	0.86	0.78	0.82	0.69	0.86	0.65
	$\tau_2$ [ns]	103	52	48	58	84	68
	b	0.61	0.58	0.54	0.62	0.62	0.76
	$\tau_{Av}$ [ns]	<b>139</b>	<b>72</b>	<b>74</b>	<b>67</b>	<b>111</b>	<b>62</b>
<b>20 min</b>	$A_1$ (rel.)	0.20	0.23	0.17	0.21	0.14	0.37
	$\tau_1$ [ns]	73	45	30	37	55	32
	$A_2$ (rel.)	0.80	0.77	0.83	0.79	0.86	0.63
	$\tau_2$ [ns]	165	76	70	62	77	59
	b	0.73	0.65	0.62	0.73	0.60	0.73
	$\tau_{Av}$ [ns]	<b>175</b>	<b>90</b>	<b>89</b>	<b>68</b>	<b>108</b>	<b>57</b>