Supporting Information for "Imaging Excited State Dynamics in Layered 2D Perovskites with Transient Absorption Microscopy"

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I. Tabulated Fitting Results

Parameters obtained from fitting absorbance lineshapes to Equation 1 are presented in the tables below. Fitting parameters are obtained after the broad continuum contribution has been successfully isolated and removed, leaving only the Gaussian lineshapes associated with the quantum well resonances. Parameters presented in Table S1 are obtained after averaging together absorbance profiles across multiple samples with depths differing by no more than 20nm.

Table S1. Spectral fitting parameters for average absorbance lineshapes.

Fitting Parameters	Full Thickness	115 nm	160 nm	180 nm	220 nm	245 nm	350 nm	370 nm
c_1	0.230	0.240	0.220	0.197	0.169	0.132	0.091	0.073
ω_1 (cm^{-1})	19400	19400	19400	19400	19300	19300	19400	19500
d_1 (cm^{-1})	800	717	723	633	650	601	679	829
c_2	0.263	0.267	0.232	0.229	0.213	0.179	0.087	0.082
ω_2 (cm^{-1})	17600	17600	17600	17600	17600	17600	17600	17700
d_2 (cm^{-1})	600	581	600	549	593	586	681	697
c_3	0.181	0.125	0.107	0.097	0.082	0.076	0.055	0.053
ω_3 (cm^{-1})	16400	16400	16400	16400	16400	16400	16400	16400
d_3 (cm^{-1})	565	544	522	508	650	568	829	903

Table S2. Spectral fitting parameters for Sample 1.

Fitting Parameters	Full Thickness	110 nm	170 nm	220 nm	360 nm
c_1	0.242	0.244	0.233	0.170	0.078

ω_1 (cm^{-1})	19400	19400	19400	19300	19400
d_1 (cm^{-1})	868	713	735	652	832
c_2	0.273	0.277	0.248	0.214	0.082
ω_2 (cm^{-1})	17600	17600	17600	17600	17600
d_2 (cm^{-1})	636	595	600	600	710
c_3	0.180	0.131	0.102	0.083	0.059
ω_3 (cm^{-1})	16400	16400	16400	16400	16400
d_3 (cm^{-1})	560	547	540	640	799

Table S3. Spectral fitting parameters for Sample 2.

Fitting Parameters	Full Thickness	120 nm	160 nm	240 nm	345 nm
c_1	0.222	0.244	0.195	0.112	0.099
ω_1 (cm^{-1})	19500	19400	19400	19300	19400
d_1 (cm^{-1})	1060	740	759	704	697
c_2	0.250	0.262	0.196	0.163	0.092
ω_2 (cm^{-1})	17600	17600	17600	17600	17600
d_2 (cm^{-1})	626	588	620	616	702
c_3	0.177	0.117	0.090	0.082	0.059
ω_3 (cm^{-1})	16400	16400	16400	16400	16300
d_3 (cm^{-1})	578	516	597	750	790

Table S4. Spectral fitting parameters for Sample 3.

Fitting Parameters	Full Thickness	160 nm	240 nm	250 nm	355 nm
c_1	0.219	0.243	0.162	0.132	0.080
ω_1 (cm^{-1})	19400	19400	19300	19300	19400
d_1 (cm^{-1})	941	710	580	577	640
c_2	0.274	0.262	0.200	0.159	0.076

ω_2 (cm^{-1})	17600	17600	17600	17600	17600
d_2 (cm^{-1})	608	527	600	546	614
c_3	0.182	0.110	0.083	0.068	0.054
ω_3 (cm^{-1})	16400	16400	16400	16400	16400
d_3 (cm^{-1})	527	500	522	594	712

Table S5. Spectral fitting parameters for Sample 4.

Fitting Parameters	**Full Thickness**	**150 nm**	**180 nm**	**250 nm**	**380 nm**
c_1	0.207	0.214	0.201	0.111	0.066
ω_1 (cm^{-1})	19500	19400	19400	19300	19400
d_1 (cm^{-1})	957	716	635	554	759
c_2	0.277	0.248	0.232	0.175	0.076
ω_2 (cm^{-1})	17600	17600	17600	17600	17600
d_2 (cm^{-1})	607	566	564	576	658
c_3	0.181	0.125	0.094	0.071	0.049
ω_3 (cm^{-1})	16400	16400	16400	16400	16400
d_3 (cm^{-1})	527	505	487	540	850

II. Results of Milling Experiments on Individual Samples

The figures below summarize the data collected for 4 individual milling samples.

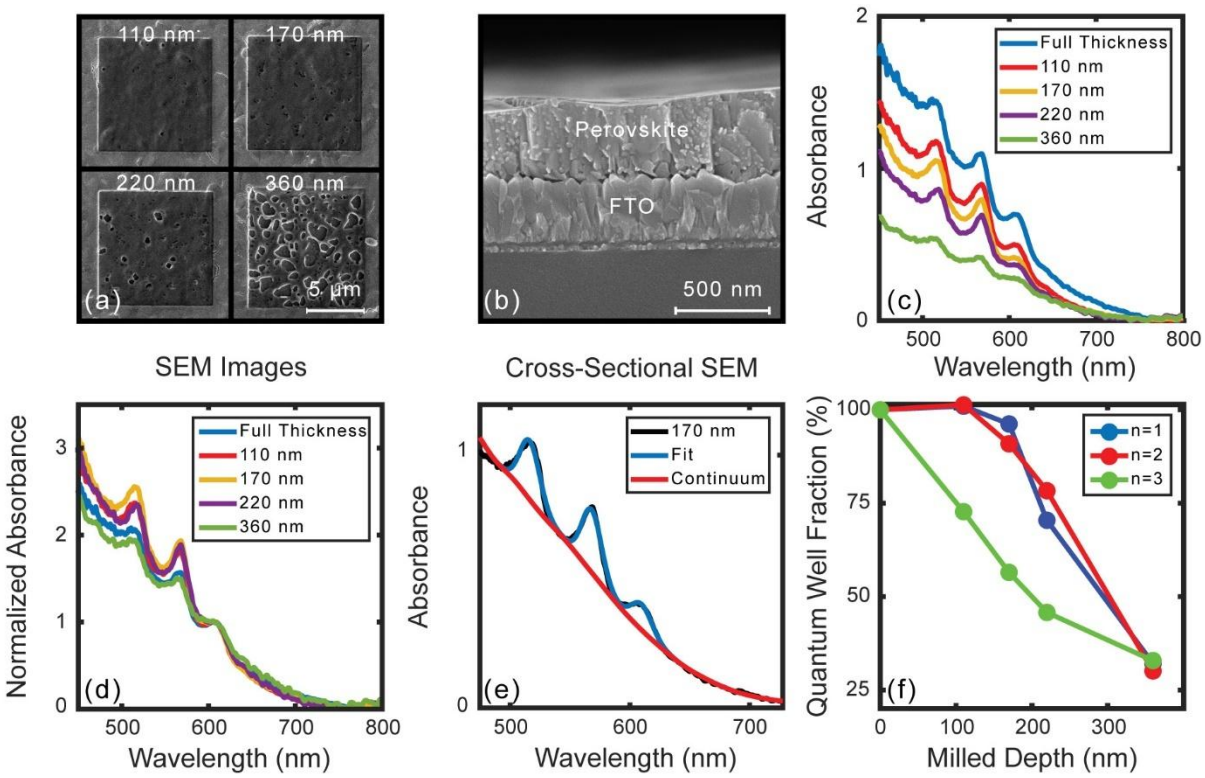

Figure S1. *Summary of milling results for Sample 1. (a) SEM images of four milled regions with indicated depths, as determined by AFM. (b) Cross-sectional SEM image showing the full thickness of the layered perovskite film. (c) Absorbance lineshapes for the full thickness of Sample 1 and milled regions. (d) Absorbance spectra are normalized to the n=3 excitonic resonance at approximately 610 nm. (e)Representative fit of the continuum and quantum well absorbance lineshapes. (f) The change in quantum well concentration as a function of milling depths. Concentrations above 100% result from a combination of experimental error and film heterogeneity.*

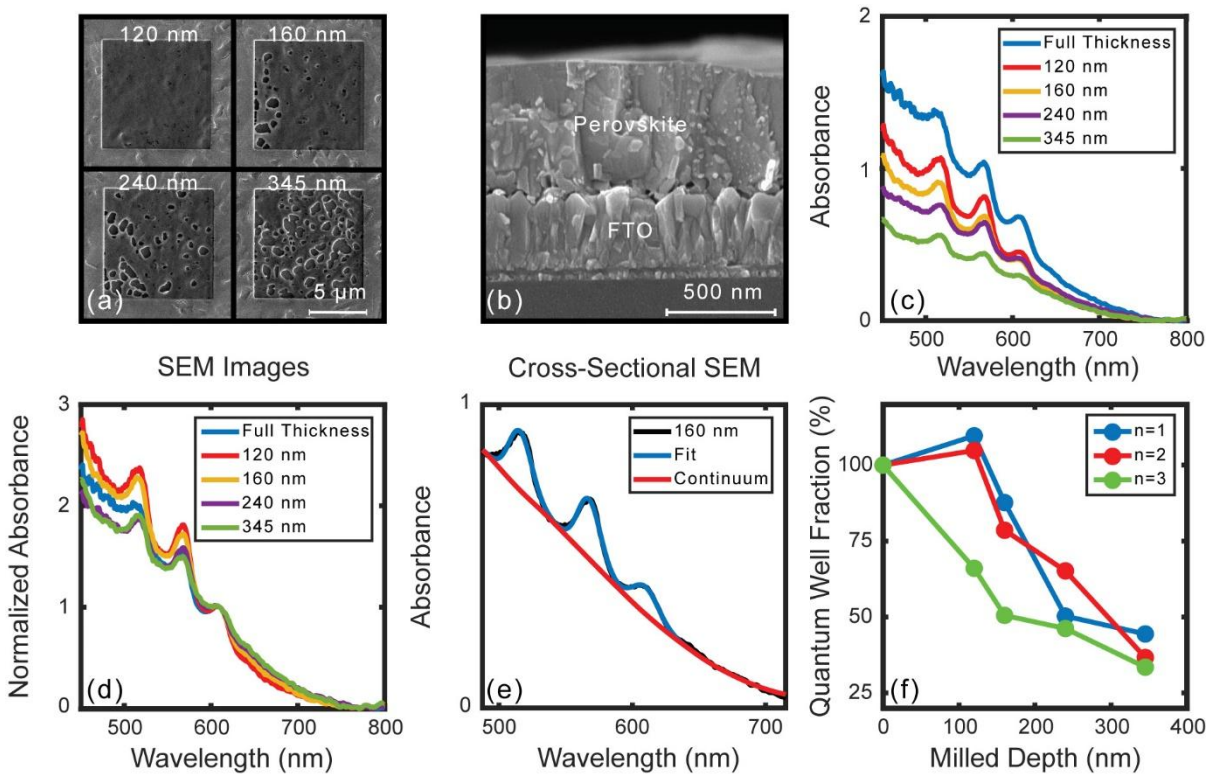

Figure S2. *Summary of milling results for Sample 2. (a) SEM images of four milled regions with indicated depths, as determined by AFM. (b) Cross-sectional SEM image showing the full thickness of the layered perovskite film. (c) Absorbance lineshapes for the full thickness of Sample 2 and milled regions. (d) Absorbance spectra are normalized to the n=3 excitonic resonance at approximately 610 nm. (e)Representative fit of the continuum and quantum well absorbance lineshapes. (f) The change in quantum well concentration as a function of milling depths. Concentrations above 100% result from a combination of experimental error and film heterogeneity.*

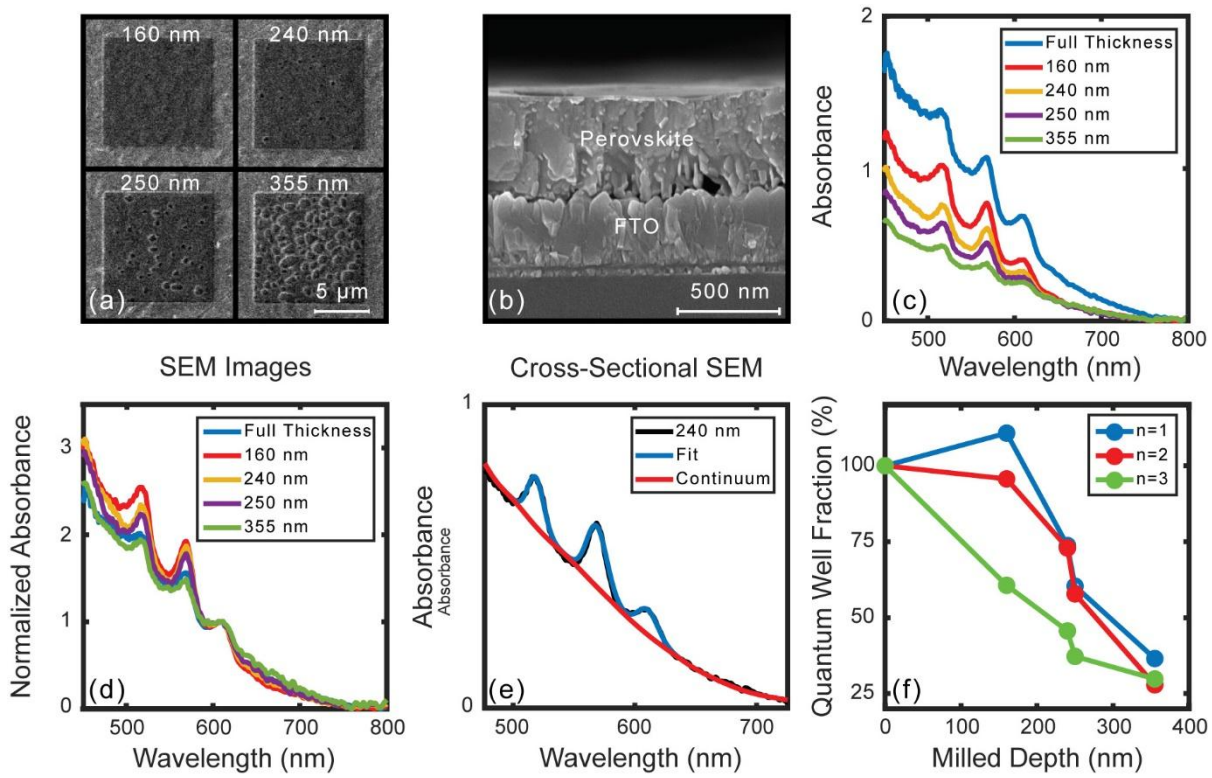

Figure S3. *Summary of milling results for Sample 3. (a) SEM images of four milled regions with indicated depths, as determined by AFM. (b) Cross-sectional SEM image showing the full thickness of the layered perovskite film. (c) Absorbance lineshapes for the full thickness of Sample 3 and milled regions. (d) Absorbance spectra are normalized to the n=3 excitonic resonance at approximately 610 nm. (e)Representative fit of the continuum and quantum well absorbance lineshapes. (f) The change in quantum well concentration as a function of milling depths. Concentrations above 100% result from a combination of experimental error and film heterogeneity.*

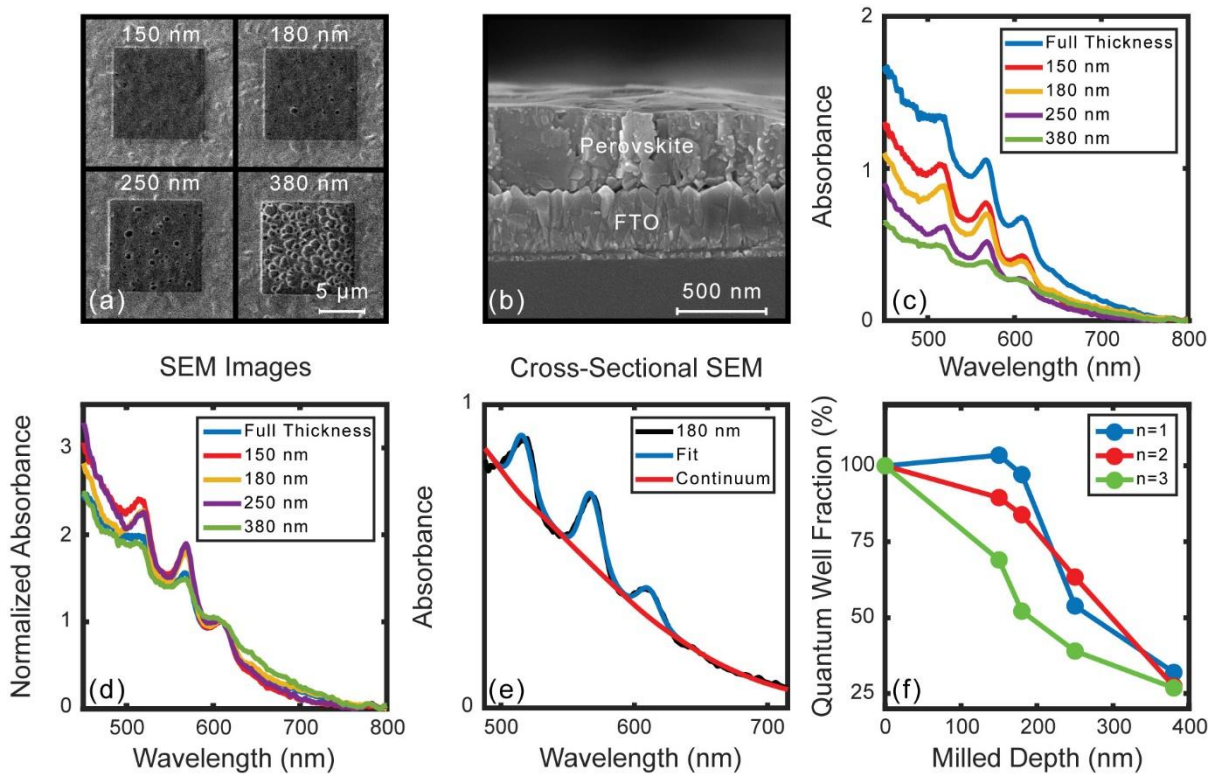

Figure S4. *Summary of milling results for Sample 4. (a) SEM images of four milled regions with indicated depths, as determined by AFM. (b) Cross-sectional SEM image showing the full thickness of the layered perovskite film. (c) Absorbance lineshapes for the full thickness of Sample 4 and milled regions. (d) Absorbance spectra are normalized to the n=3 excitonic resonance at approximately 610 nm. (e)Representative fit of the continuum and quantum well absorbance lineshapes. (f) The change in quantum well concentration as a function of milling depths. Concentrations above 100% result from a combination of experimental error and film heterogeneity.*

III. AFM Depth Profiles of Individual Samples

The figures below summarize the AFM depth profiles obtained from the four milled regions of 4 individual samples.

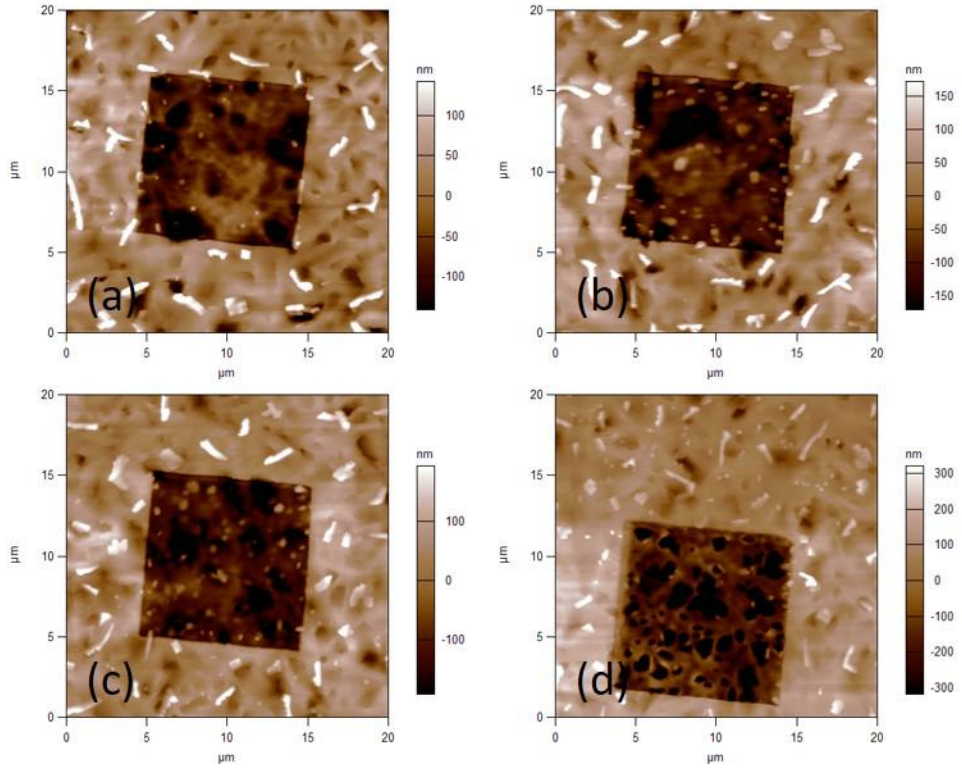

Figure S5. *AFM depth profiles for Sample 1 demonstrate average milling depths of (a) 110, (b) 170, (c) 220, and (d) 360 nm.*

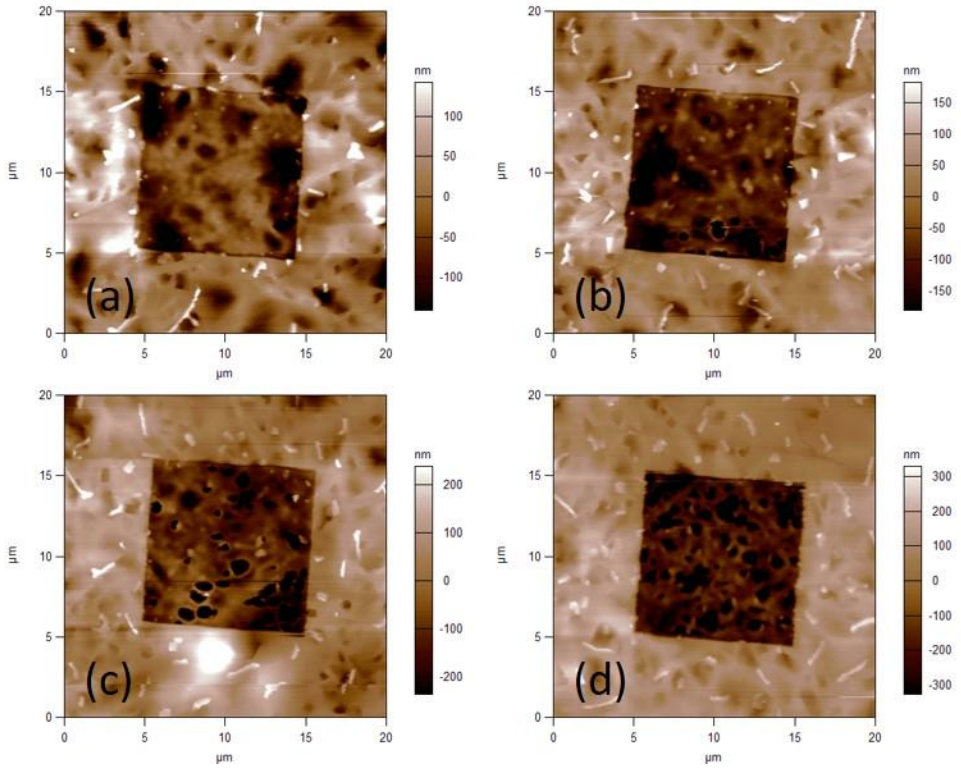

Figure S6. *AFM depth profiles for Sample 2 demonstrate average milling depths of (a) 120, (b) 160, (c) 240, and (d) 345 nm.*

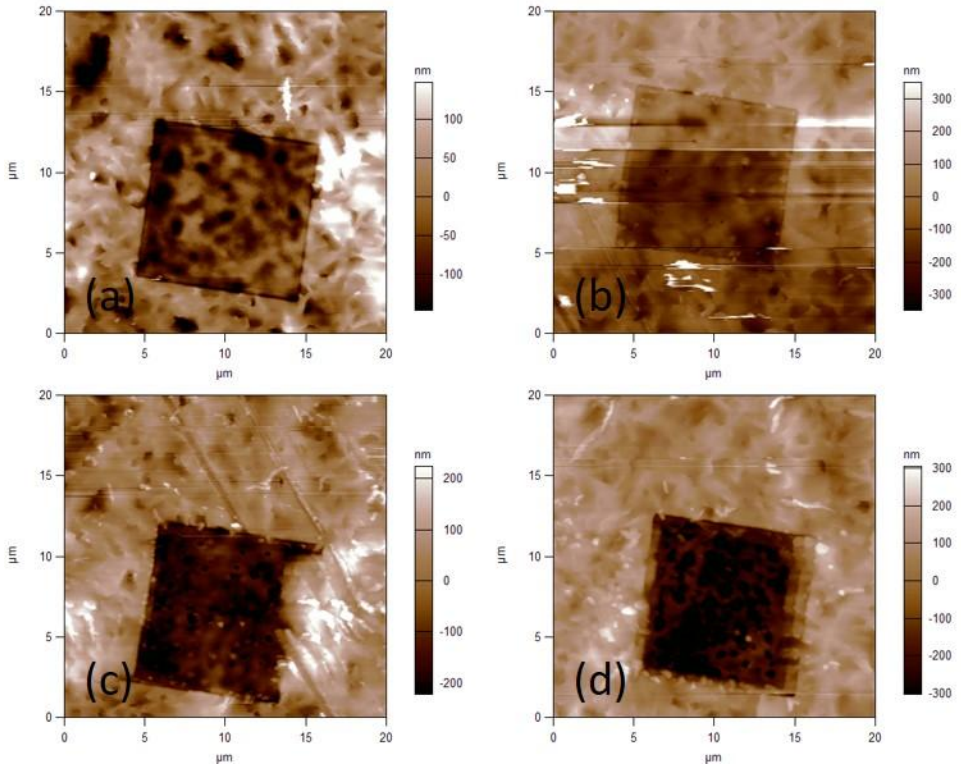

Figure S7. *AFM depth profiles for Sample 3 demonstrate average milling depths of (a) 160, (b) 240, (c) 250, and (d) 355 nm.*

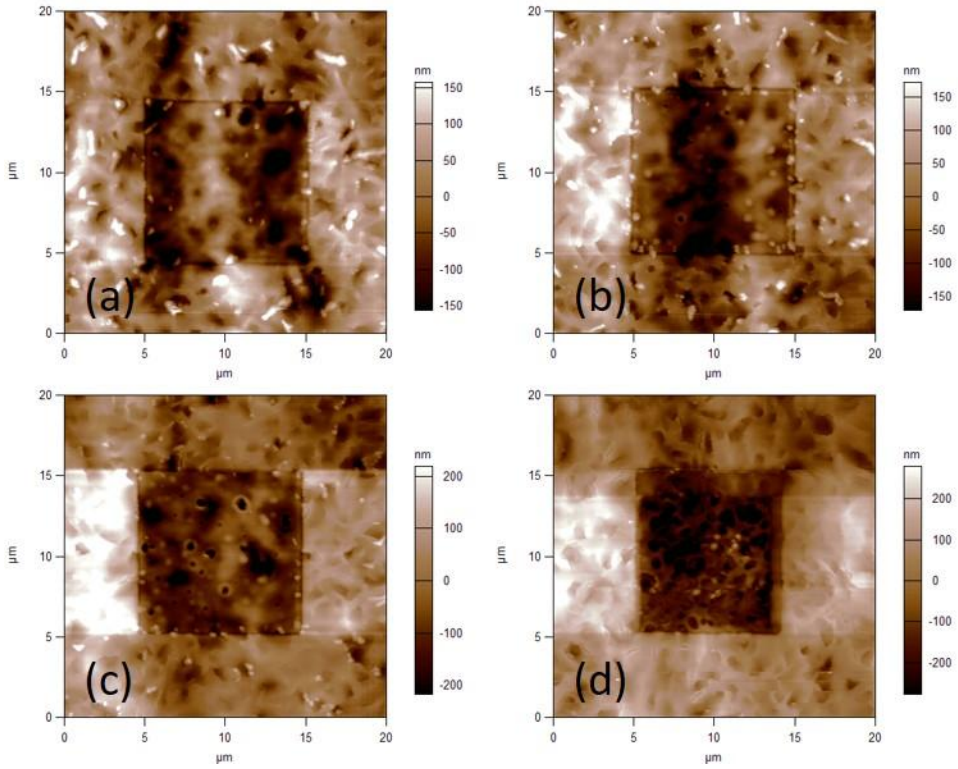

Figure S8. *AFM depth profiles for Sample 4 demonstrate average milling depths of (a) 150, (b) 180, (c) 250, and (d) 380 nm.*

IV. Linear Absorption Spectra of 2D Perovskite Single Crystals

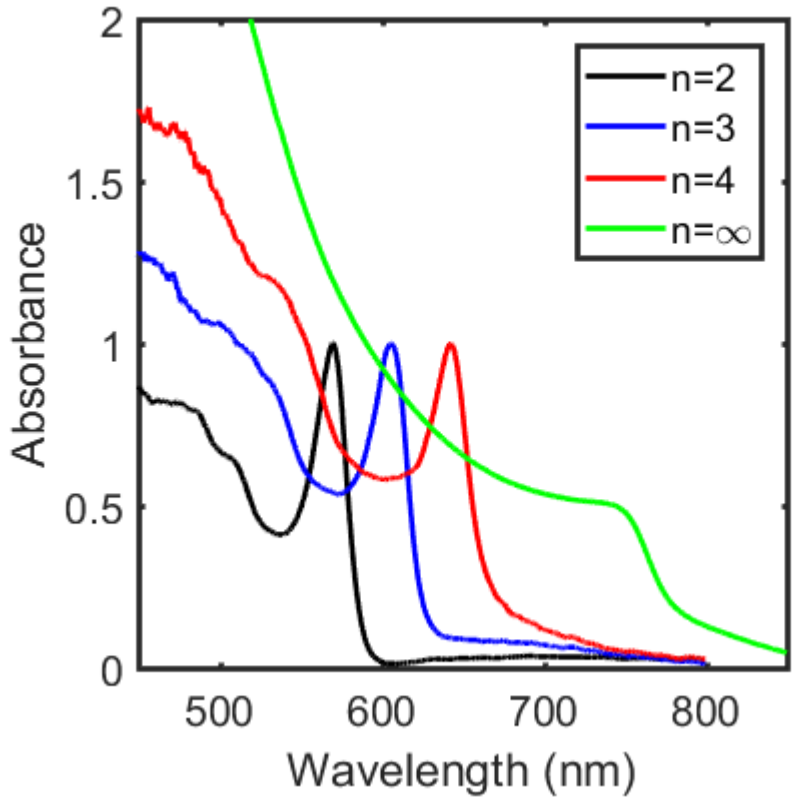

Figure S9. *Linear absorbance spectra obtained for perovskite single crystals.*