Supplementary Information for

Anisotropic Carrier Dynamics and Laser-Fabricated Luminescent Patterns on Oriented Single-Crystal Perovskite Wafers

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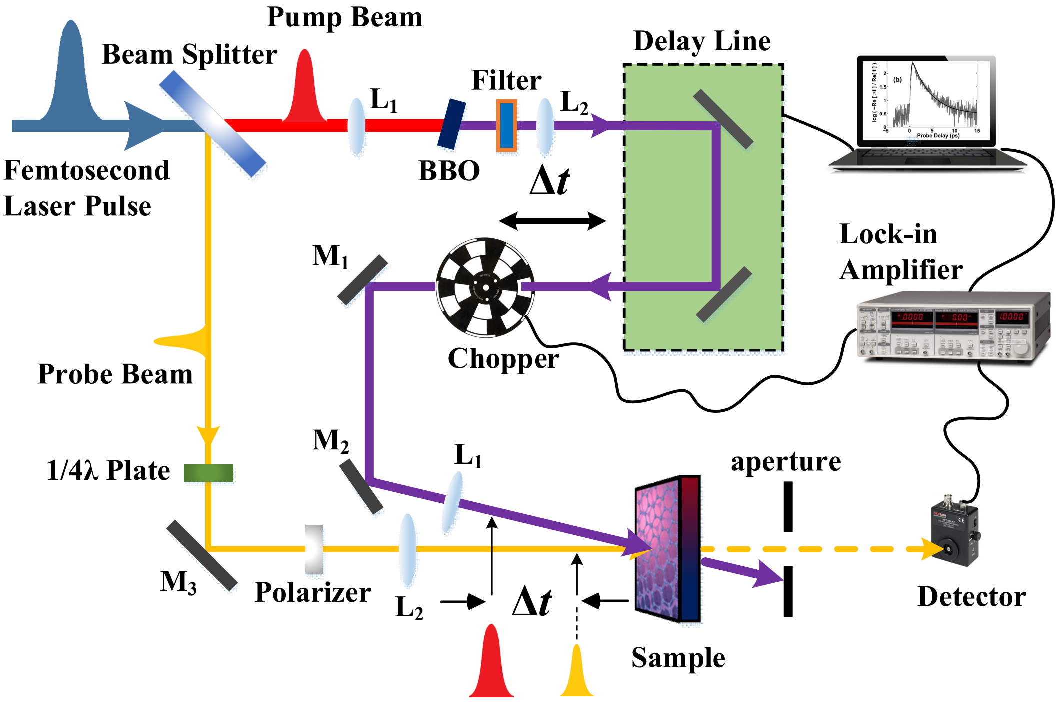

Figure S1. Schematic illustration of the angle-resolved transient transmission measurement based on ultrafast time-resolved pump-probe technique. The polarization of the probe light is modulated by a 1/4 λ wave plate and a polarizer.

Table S1. Pump fluence-dependent transient transmittance with pump and probe polarization fixed along 0° and 90°. The $|\Delta T/T_0|$ of the MAPbBr$_3$ (001) wafer is fitted by a double-exponential model. The decay parameters of carrier relaxation dynamics are tabulated in **Table S1.**

Fitting Function	$\Delta T/T_0 = A_1$*exp($-t/\tau_1$) + A_2*exp($-t/\tau_2$) + A_0					
Pump Fluence (μJ/cm^2)	71	142	213	284	355	426
A_1	4.84522E-5 ± 8.42105E-7	1.1273E-4 ± 1.34055E-6	1.77473E-4 ± 1.69408E-6	2.49837E-4 ± 2.41161E-6	3.17259E-4 ± 3.00205E-6	3.34575E-4 ± 2.71281E-6
τ_1 (ps)	14.83228 ± 0.49197	8.62977 ± 0.2055	6.70164 ± 0.12147	5.17426 ± 0.08929	4.93 ± 0.08264	5.05273 ± 0.07347
A_2	3.83089E-5 ± 7.40399E-7	6.47769E-5 ± 9.59491E-7	8.65664E-5 ± 9.57498E-7	1.06843E-4 ± 1.1553E-6	1.2565E-4 ± 1.41204E-6	1.31599E-4 ± 1.32792E-6
τ_2 (ps)	105.19739 ± 3.09251	74.89678 ± 1.43611	68.87816 ± 1.01044	57.7027 ± 0.78406	55.11021 ± 0.76497	54.91648 ± 0.6767

Table S2. Probe polarization dependence fitting for carrier relaxation dynamics in $MAPbBr_3$ (100) wafer. (pump laser fluence: 71 μJ/cm^2)

Fitting Function	$\Delta T/T_0 = A_1$*exp($-t/\tau_1$) + A_2*exp($-t/\tau_2$) + A_0							
Polarization	0°	15°	30°	60°	120°	150°	165°	180°
A_1	7.2048E-5 ± 1.97632E-6	3.5567E-5 ± 1.03273E-6	3.7847E-5 ± 1.09701E-6	2.5256E-5 ± 1.27872E-6	2.5235E-5 ± 1.07597E-6	3.7847E-5 ± 3.97675E-6	3.5567E-5 ± 2.05784E-6	8.0007E-5 ± 1.17381E-6
τ_1 (ps)	14.0012 ± 0.32934	8.0809 ± 0.45067	4.8295 ± 0.42119	4.2432 ± 0.63425	4.2352 ± 0.48635	4.8295 ± 0.38642	8.0809 ± 0.78842	13.928 ± 0.55661
A_2	4.6097E-5 ± 8.37473E-7	3.004E-5 ± 7.07383E-7	2.3597E-5 ± 6.80884E-7	1.8085E-5 ± 9.68453E-7	1.8076E-5 ± 5.99884E-7	2.3597E-5 ± 7.27026E-7	3.004E-5 ± 7.55944E-7	3.7474E-5 ± 8.20683E-7
τ_2 (ps)	116.2476 ± 3.38474	92.9825 ± 5.16539	87.8235 ± 4.68596	68.9952 ± 8.62745	68.9924 ± 7.08953	87.8257 ± 4.89247	92.9858 ± 12.01956	115.2447 ± 5.67865

Table S3. Probe polarization dependence fitting for carrier relaxation dynamics in $MAPbBr_3$ (110) wafer. (pump laser fluence: 71 μJ/cm^2)

Fitting Function	$\Delta T/T_0 = A_1$*exp($-t/\tau_1$) + A_2*exp($-t/\tau_2$) + A_0							
Polarization	0°	45°	90°	135°	180°	225°	270°	315°
A_1	1.0053E-4 ± 3.8756E-6	1.14681E-4 ± 5.18882E-6	1.14137E-4 ± 5.92581E-6	1.04129E-4 ± 6.6789E-6	1.0343E-4 ± 7.00502E-6	1.11756E-4 ± 1.04812E-5	1.07003E-4 ± 1.17891E-5	1.16032E-4 ± 1.1798E-5
τ_1 (ps)	6.72274 ± 0.33702	6.85274 ± 0.3108	6.61291 ± 0.33241	6.79207 ± 0.21803	6.94306 ± 0.50863	6.76792 ± 0.41433	6.54351 ± 0.35064	6.67874 ± 0.32694
A_2	2.00474E-5 ± 9.29897E-7	2.12277E-5 ± 1.06459E-6	2.04683E-5 ± 1.05788E-6	1.85936E-5 ± 7.78636E-7	1.94785E-5 ± 1.33657E-6	1.9728E-5 ± 1.48011E-6	2.01666E-5 ± 1.32997E-6	1.83313E-5 ± 1.19156E-6
τ_2 (ps)	85.21401 ± 6.1192	87.25635 ± 5.08631	83.12434 ± 6.00969	86.42331 ± 4.629	82.87642 ± 10.50368	87.13354 ± 8.61594	84.31272 ± 6.41169	88.32177 ± 8.04558

Table S4. Probe polarization dependence fitting for carrier relaxation dynamics in $MAPbBr_3$ (111) wafer. (pump laser fluence: 71 μJ/cm^2)

Fitting Function	$\Delta T/T_0 = A_1$*exp($-t/\tau_1$) + A_2*exp($-t/\tau_2$) + A_0								
Polarization	0°	15°	30°	60°	90°	135°	150°	165°	180°
A_1	4.0884E-5 ± 2.71828E-6	3.6723E-5 ± 1.96143E-6	1.9757E-5 ± 2.54963E-6	2.8919E-5 ± 2.13805E-6	2.3313E-5 ± 1.78655E-6	2.9884E-5 ± 2.05173E-6	2.5281E-5 ± 2.62371E-6	3.2678E-5 ± 2.07646E-6	4.4969E-5 ± 2.57421E-6
τ_1 (ps)	11.0012 ± 0.62306	10.2182 ± 0.77969	8.5668 ± 1.52384	6.7205 ± 1.10726	7.4085 ± 1.62286	7.0256 ± 0.90435	9.4647 ± 1.35111	10.9964 ± 1.12919	10.8177 ± 0.87957
A_2	2.4415E-5 ± 1.23715E-6	1.4938E-5 ± 1.15786E-6	2.2855E-5 ± 1.93386E-6	1.4885E-5 ± 8.79473E-7	1.6698E-5 ± 1.45519E-6	1.9051E-5 ± 9.91409E-7	1.8682E-5 ± 1.62644E-6	1.5565E-5 ± 1.18487E-6	1.5929E-5 ± 1.17194E-6
τ_2 (ps)	125.49632 ± 7.39257	94.54635 ± 10.79514	85.2072 ± 4.85921	86.69317 ± 15.35244	78.55145 ± 15.49168	85.58077 ± 13.11174	82.49376 ± 8.58629	103.9589 ± 14.42511	124.09893 ± 11.97071

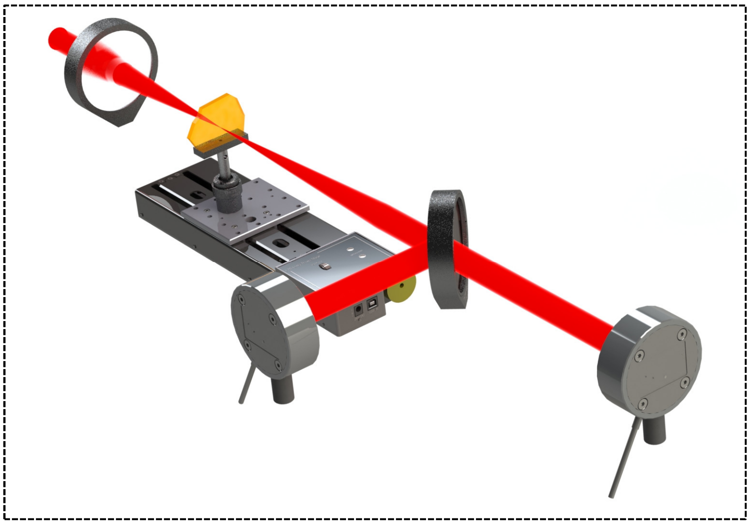

Figure S2. Schematic illustration of the transmission open-aperture *Z*-scan technique. The experiment is carried by moving the sample back-and-forth from the focal point of the pulsed laser beam along its optical axis (defined as the *z*-axis).

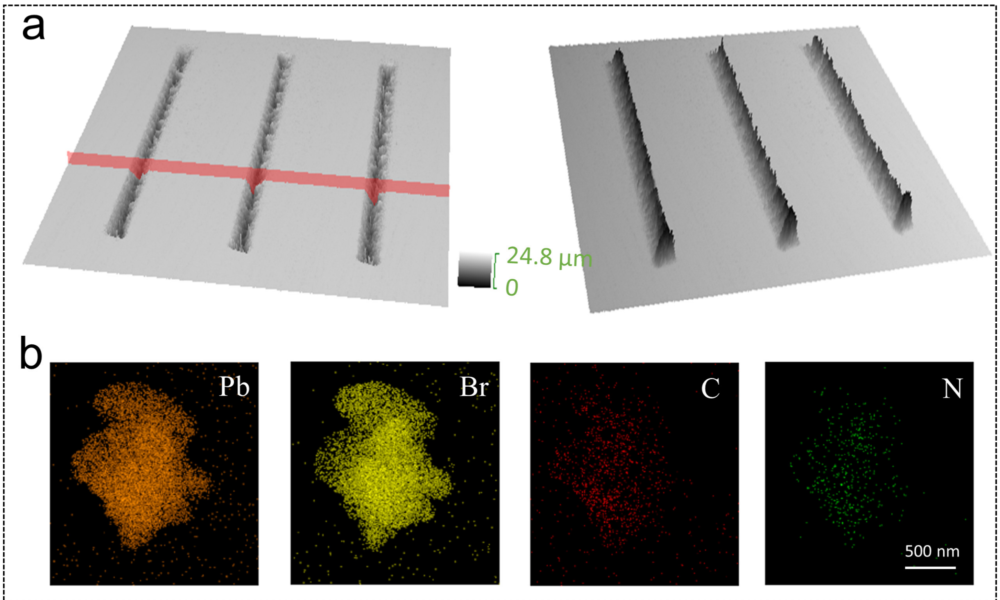

Figure S3. The process homogeneity and chemical stability of laser-fabricated luminescent micro-patterns on oriented wafers. (**a**) The front-view and back-view height profile images of laser scanning confocal microscopy (LSCM) indicates uniform processing depth and laser stability under the same processing parameters [laser power: 5 mW, defocus: 470 μm and scanning speed: 0.1 mm s^{-1}]. (**b**) EDS mapping images show that Pb, Br, C and N elements are uniformly distributed in the processed regions of $MAPbBr_3$.

Table S5. The element content of the processed regions of $MAPbBr_3$.

Element	Pb M	Br L	C K	N K	Totals
Weight %	42.34	49.02	6.38	2.27	100
Atomic %	13.54	40.60	35.14	10.73	100

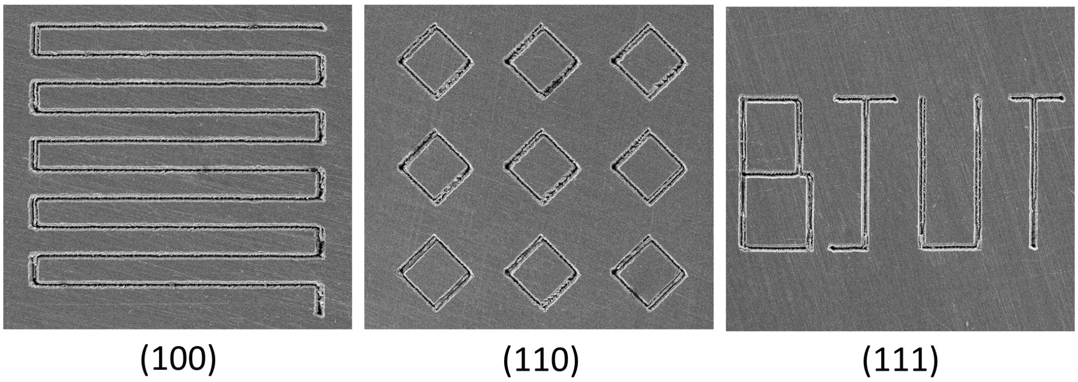

Figure S4. The scanning electron microscope (SEM) images of femtosecond laser processed scalable patterns on three $MAPbBr_3$ wafers. [laser power: 5 mW, defocus: 470 μm and scanning speed: 0.1 mm s^{-1}]

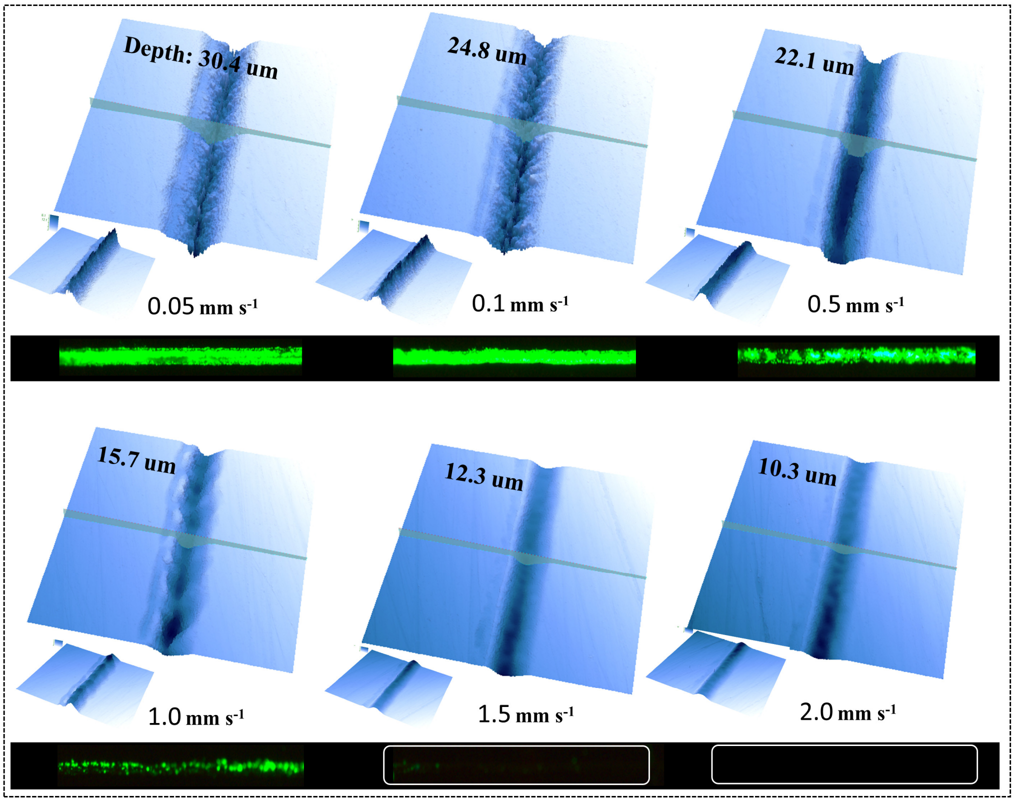

Figure S5. Effect of processing scanning speed on luminescence. The front-view and back-view colored 3D profile images of LSCM and corresponding fluorescent microscopy (FM) images at different processing scanning speeds indicate that the scanning speed of 0.1 mm s^{-1} corresponds to the strongest fluorescence emission. [laser power: 5 mW, defocus: 470 μm]

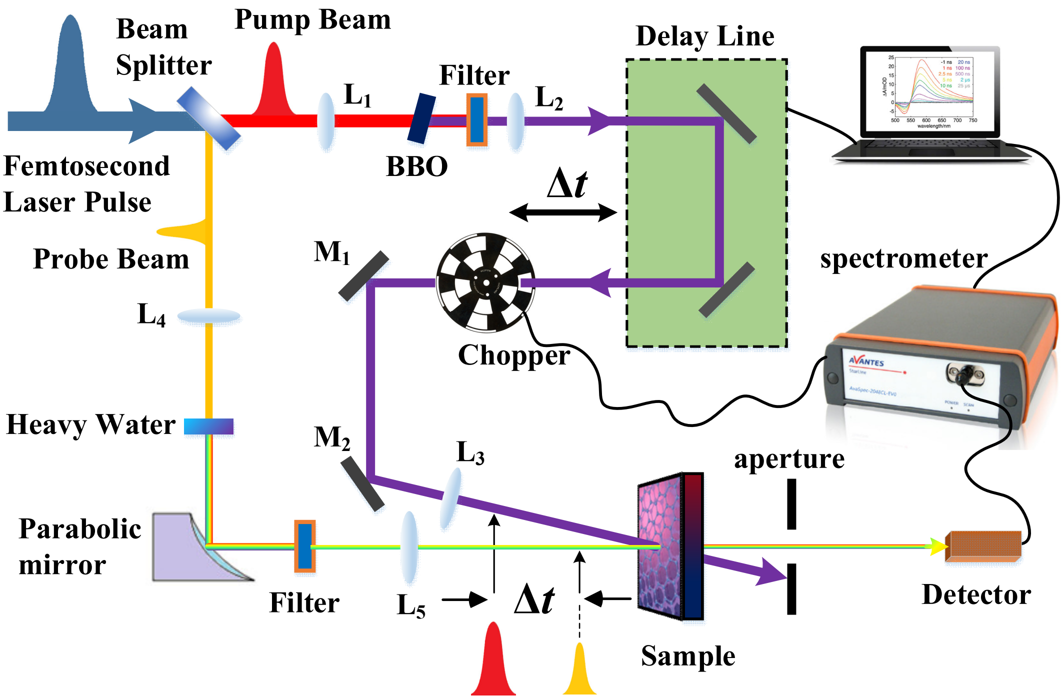

Figure S6. Schematic illustration of the transient absorption spectrum (TAS). Transient absorption spectroscopy measurements were performed using the aforementioned single-wavelength transient transmittance system in **Figure S1**. In contrast, the probe light is replaced by a supercontinuum of about 400 ~ 960 nm generated by focusing the original 800 nm femtosecond pulse into the heavy water.

Table S6. Probe wavelength dependence fitting for carrier relaxation dynamics in unprocessed MAPbBr3 (100) wafer. The wavelength of 556 nm corresponds to the ground state bleaching dynamics, and the wavelength of 703 nm corresponds to the trap state dynamics. (pump laser power: 400 μW)

Fitting Function	$\Delta T/T_0 = A_1$*exp($-t/\tau_1$) + A_0	$\Delta T/T_0 = A_1$*exp($-t/\tau_1$) + A_2*exp($-t/\tau_2$) + A_0	$\Delta T/T_0 = A_1$*exp($-t/\tau_1$) + A_0	$\Delta T/T_0 = A_1$*exp($-t/\tau_1$) + A_2*exp($-t/\tau_2$) + A_0
Wavelength (nm)	556	678	703	740
A_1	-0.01576 ± 6.03882E-4	0.0918 ± 0.01434	0.00846 ± 4.80615E-4	0.17392 ± 0.03566
τ_1 (ps)	17.77671 ± 1.17046	2.12877 ± 0.22979	5.9888 ± 0.33367	1.69659 ± 0.19474
A_2		0.00991 ± 0.00106		0.01309 ± 0.00164
τ_2 (ps)		23.78674 ± 3.83041		18.46666 ± 2.79338

Table S7. Probe wavelength dependence fitting for carrier relaxation dynamics in processed MAPbBr3 (100) wafer. The relaxation dynamics at the wavelength of 703nm are similar to those at 678nm and 740nm, indicating that the surface trap states located near the 703 nm energy level before processing disappear. (pump laser power: 400 μW)

Fitting Function	$\Delta T/T_0 = A_1*\exp(-t/\tau_1) + A_0$	$\Delta T/T_0 = A_1*\exp(-t/\tau_1) + A_2*\exp(-t/\tau_2) + A_0$	$\Delta T/T_0 = A_1*\exp(-t/\tau_1) + A_2*\exp(-t/\tau_2) + A_0$	$\Delta T/T_0 = A_1*\exp(-t/\tau_1) + A_2*\exp(-t/\tau_2) + A_0$
Wavelength (nm)	559	678	703	740
A_1	-0.01579 ± 6.93877E-4	0.05567 ± 0.00914	0.04345 ± 0.00771	0.06488 ± 0.0117
τ_1 (ps)	17.91842 ± 1.44724	1.86811 ± 0.28669	1.97062 ± 0.35942	1.95781 ± 0.36298
A_2		0.00822 ± 0.0015	0.00703 ± 0.00165	0.00935 ± 0.00253
τ_2 (ps)		16.76311 ± 3.41146	15.64508 ± 3.75102	15.30547 ± 4.16081