

Name and DOI of manuscript: Excited-state dynamics of MAPbBr₃: Coexistence of excitons and free charge carriers at ultrafast times, <https://doi.org/10.1021/acs.jpcc.3c08509>

Data availability

Data measured by the Banerji group in agreement with data management policies (SNF, Horizon 2020) and shown in the main manuscript is made publicly available in the BORIS repository of the University of Bern. For raw data, S.I. data and data acquired by collaborators, please contact the authors (available upon request).

Data acquisition: Details on the methods of data acquisition are described in the above manuscript and the corresponding S.I.

Data and analysis:

Figure 1

(a) SEM image of the MAPbBr₃ film. The inset displays a higher magnification image. (b) Normalised absorption coefficient spectrum. The Gaussian analysis of the excitonic peak is included with a dotted line. (c) Time integrated PL spectra obtained with low (10^{16} cm⁻³) and high ($3 \cdot 10^{18}$ cm⁻³) excitation densities. The Gaussians used for fitting the results are also depicted. Excitation at 450 nm was used. (d) Integrated PL intensity detected at the PL maximum, once with increasing (red circles) and once with decreasing (blue circles) the excitation carrier density.

Figure 1 data: b) Figure1b.txt

c) Figure1c.txt

d) Figure1d.txt

Person who measured: Nikos Droseros

Reference to lab book: Notebook Number 4 (page 12-30)

Figure 2

(a) Photoconductivity spectra recorded at different time delays after excitation at 480 nm and 540 nm with a photoexcited carrier density of $6.3 \cdot 10^{17}$ cm⁻³, which is in the linear regime. Both the real (squares) and the imaginary (open circles) parts are displayed. (b) Normalized THz dynamics (ΔE_{THz}) and TA (GSB) dynamics (ΔA_{TA}) with 480 nm and 540 nm excitation. The THz dynamics at early times are included in the inset.

Figure 2 data: a) Figure2a.txt

b) Figure2b.txt

Person who measured: Demetra Tsokkou

Reference to lab book: Notebook Number 4 (120619 – 030719)

Figure 3

(a) TA spectra after excitation at 480 nm with a photogenerated carrier density of $1.2 \cdot 10^{17}$ cm⁻³. The steady state absorption spectrum (red), and the pump pulse width used (blue) are also

shown, with their amplitude being arbitrary. (b) Decay-associated spectra of the MAPbBr₃ film excited at 480 nm. (c) Normalized TA dynamics at 3 ps probed at different wavelengths in the GSB 1; the arrows point towards longer probed wavelengths. (d) Band diagram of the MAPbBr₃ film around the Γ -point along with the detected optical transitions.

Figure 3 data: a) Figure3a.txt

b) Figure3c.txt

Person who measured: Nikos Droseros

Reference to lab book: Notebook Number 4 (10.05-17.0519)

Figure 4

(a) TA spectra following excitation with 540 nm at selected times and a photoexcited carrier density of $4.3 \cdot 10^{16} \text{ cm}^{-3}$. The steady state absorption spectrum (red), and the pump pulse width used (green) are also shown, with their amplitude being arbitrary. (b) Decay-associated spectra of the MAPbBr₃ film excited at 540 nm. The inset in Figure 4b displays TA dynamics at various probed wavelengths along with their respective fits derived from the multiexponential global analysis. (c) and (d) TA spectra with 480 nm (blue line) and 540 nm (red line) excitations, and their difference spectra (black line) detected at (c) 1 ps and (d) 1980 ps. Prior to subtraction, the TA spectra for both excitations were normalised at 505 nm.

Figure 4 data: a) Figure4a.txt

b) Figure4b.txt

c) Figure4c.txt

d) Figure4d.txt

Person who measured: Nikos Droseros

Reference to lab book: Notebook Number 4 (10.05-17.0519)

Figure 5

TA spectra under various excitation densities with 480 nm excitation, detected (a) at 200 fs and (b) at 1980 ps after excitation. TA spectra under various excitation densities with 540 nm excitation, detected (c) at 200 fs and (d) at 1980 ps after excitation. The inset in Figure 5d displays the energy difference between the positive excitonic peak and that in the steady-state absorption spectrum. The analysis of the data with a linear function is included in the same graph. Here, a logarithmic scale is used for the x-axis.

Figure 5 data: a) Figure5a.txt

b) Figure5b.txt

c) Figure5c.txt

d) Figure5d.txt

Person who measured: Nikos Droseros

Reference to lab book: Notebook Number 4 (10.05-17.0519)