Photo-induced hot carrier cooling in two-dimensional perovskite single crystal

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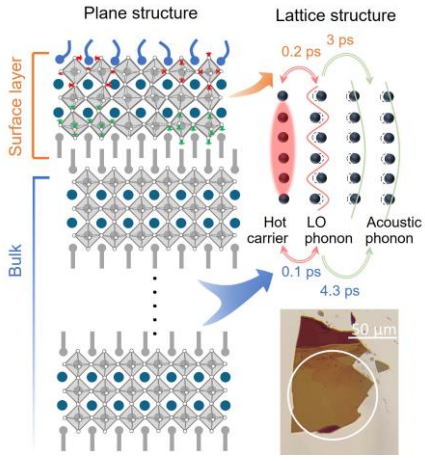
Hot carrier solar cell (HCSC) is a promising concept for overcoming Shockley-Queisser limit[1-2]. Here we investigate the potential of two-dimensional (2D) organic-inorganic hybrid perovskites [$(BA)_2(MA)_2Pb_3I_{10}$, BA = Butylammonium, MA = Methylammonium] as a building block in HCSC. To reveal the HC cooling mechanisms in 2D perovskite single crystal, we apply two complementary ultrafast spectroscopy techniques - transient absorption(TA) and time-resolved two-photon photoemission (TR-2PPE). While TA results provide information about the whole bulk, TR-2PPE directly maps the hot electron distribution and its dynamics from surface layer of the single crystal. In general, both dynamics exhibit two cooling stages where the fast one is attributed to the carrier-LO (longitudinal optical) phonon scattering, while the energy redistribution from LO phonon to acoustic phonons plays a role in the slower stage. We found that hot carriers on the surface exhibit slower initial cooling and a lower starting temperature for the second stage compared to the bulk area.

Scheme 1: Schematic of HC cooling pathways in 2D perovskite single-crystal flake.



Acknowledgements
This work was supported by Swedish Research Council, KAW Foundation, Chinese Scholarship Council and NanoLund. The work was carried out in close collaboration with Technical University of Denmark, MAX IV laboratory and Synchrotron SOLEIL.

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