# Supramolecular Engineering of Hybrid Perovskite Solar Cells

<u>Jovana V. Mili</u>ć,[a] Dominik J. Kubicki,[a,b] Jeong-Hyeok Im,[a] Yang Li,[a]
Amita Ummadisingu,[a] Lyndon Emsley,[b] and Michael Graetzel[a]

[a] Laboratory of Photonics and Interfaces, EPFL, Lausanne, Switzerland
[b] Laboratory of Magnetic Resonance, EPFL, Lausanne, Switzerland

Hybrid perovskite solar cells exhibit remarkable power conversion efficiencies, while their limited stability and molecular-level engineering remain challenging.[1-3] In contrast to three-dimensional (3D) perovskites, their layered two-dimensional (2D) analogs have demonstrated promising stabilities, though at the expense of the corresponding efficiencies.[1,4-5] We demonstrate a strategy to provide stabilization without compromising the efficiency by employing multifunctional molecular modulators designed through fine-tuning noncovalent interactions complemented by structural adaptability.[2-5] These systems are devised to interact with the 3D perovskite surface in a manner uniquely assessed by solid-state NMR spectroscopy.[2-3] As a result, we obtain perovskite solar cells with superior properties and efficiencies exceeding 20% for formamidinium cesium mixed lead iodide compositions, accompanied by enhanced stabilities.[3] Moreover, extending the design into layered 2D architectures leads to stability enhancements.[4-5] This approach has been investigated using a combination of techniques complemented by solid-state NMR to unravel the design principles and exemplify the role of supramolecular engineering in advancing perovskite solar cells.
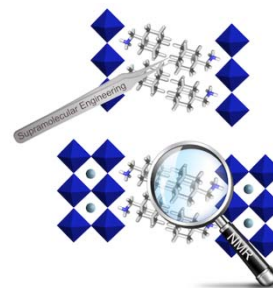
Schematic of a layered hybrid perovskite prototype

―――
[1] J. V. Milić, D. J. Kubicki, L. Emsley, M. Graetzel, *Chimia* (2019) *73*, 317. [2] M. M. Tavakoli, W. Tress, J. V. Milić, D. J. Kubicki, L. Emsley, M. Graetzel, *Energy Environ. Sci.* (2018), *11*, 3310. [3] D. Bi, X. Li, J. V. Milić*, N. Pellet, J. Luo, T. LaGrange, P. Mettraux, L. Emsley, S. M. Zakeeruddin, M. Graetzel, *Nature Commun.* (2018) *9*, 4482. [4] Y. Li, J. V. Milić*, A. Ummadisingu, J.-Y. Seo, J.-H. Im, H.-S. Kim, Y. Liu, M. I. Dar, S. M. Zakeeruddin, P. Wang, A. Hagfeldt, M. Graetzel, *Nano Lett.* (2019) *19*, 150. [5] J. V. Milić, J.-H. Im, D. J. Kubicki, A. Ummadisingu, J.-Y. Seo, Y. Li, M. A. Ruiz Preciado, M. I. Dar, S. M. Zakeeruddin, L. Emsley, M. Graetzel, *Adv. Energy Mater.* (2019) 1900284.