Topological Frenkel excitons in 2D molecular crystals

Quentin Stein[1], Siddhartha Nayak[2], Jakob Kuhlke[3], Song Jiang[1], Michelangelo Romeo[1], Fabrice Scheurer[1], Katharina Kaiser[1,3], Thomas Frederiksen[4,5], Tomáš Neuman[2] and Guillaume Schull[1]

[1]Université de Strasbourg, CNRS, IPCMS, UMR 7504, F-67000 Strasbourg, France.
[2]Institute of Physics, Czech Academy of Sciences Cukrovarnick´a 10, Prague, 16200, Czech Republic
[3]IV. Physical Institute – Solids and Nanostructures, Georg-August-Universität Göttingen, Göttingen, 37077, Germany
[4]Donostia International Physics Center, Donostia-San Sebasti´an 20018, Spain
[5]Ikerbasque, Basque Foundation for Science, E-48013 Bilbao, Spain

Controlling energy transport in molecular aggregates is a central challenge in many branches of photophysics, ranging from molecular optoelectronics to artificial light-harvesting complexes[1,2]. In organized two-dimensional (2D) molecular assemblies, theory predicts the emergence of Frenkel excitons forming continuous energy bands[3]. Here we use scanning tunneling microscopy-induced luminescence (STML) to probe the real-space wave-functions of Frenkel excitons in 2D zinc phtalocyanine molecular aggregates with sub-nanometer resolution[4]. A direct comparison between aggregates of different sizes shows a transition from quantum-dot-like systems with discrete quantum excitations to molecular crystals hosting continuous excitonic bands. We further identify an excitonic state confined to the edge of the crystal. Theoretical simulations[5] supports the emergence of this state from the momentum-resolved topology of the bulk excitonic bands. Our findings put forward 2D molecular crystals as a platform to access topologically-confined excitonic states and a promising tool to directly control energy transport through excitonic band engineering.

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