

Ultrafast Coherent Exciton Dynamics in a Series of Cofacially Stacked Perylene Bisimides

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Fundamental understanding of exciton dynamics, such as exciton (de)localization, and charge transfer processes, in functional molecular assemblies is desirable, since these photophysical phenomena strongly affect the performance of organic-material-based electronic and photonic devices. Here, we will discuss the ultrafast coherent exciton dynamics in a series of cofacially stacked perylene bisimides (PBIs). First, we present coherent exciton transport and excimer formation dynamics from the Frenkel state of PBI dimeric and oligomeric H-aggregates [1]. From the vibronic peak ratio analysis in the early-time transient fluorescence spectra obtained by femtosecond broadband fluorescence upconversion spectroscopy, the initial spatial coherence and its evolution are directly unraveled. Second, we introduce symmetry breaking charge separation dynamics via excimer intermediate state in a cyclophane bridged PBI dimer [2,3]. Based on our observation that the rise time of PBI anion and cation bands in the transient absorption spectra is equivalent to the decay time of the excimer fluorescence, we suggest that the excimer state can effectuate the charge transfer dynamics in the cofacially stacked PBI dimer. Our findings on ultrafast coherent exciton dynamics in various PBI aggregate systems will provide valuable insights into future applications in the field of molecular optoelectronic materials to achieve long-range coherent energy transfer and superb charge transfer efficiency.

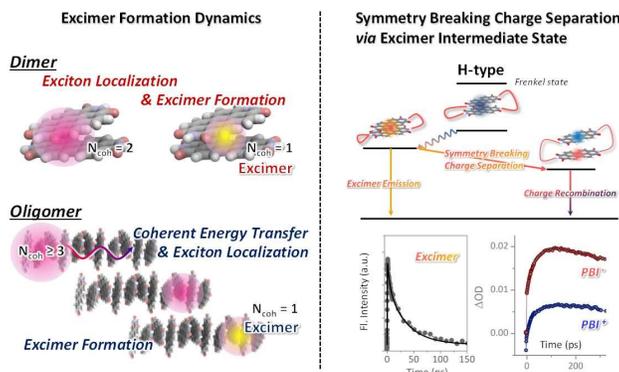


Figure 1. Ultrafast coherent exciton dynamics in PBI aggregate systems

References:

- [1] J. Sung, P. Kim, B. Fimmel, F. Würthner, and D. Kim, *Nat. Commun.* **6**, 8646 (2015).
- [2] J. Sung, A. Nowak-Król, F. Schlosser, B. Fimmel, W. Kim, D. Kim, and F. Würthner, *J. Am. Chem. Soc.* **138**, 9029 (2016).
- [3] W. Kim, A. Nowak-Król, F. Schlosser, B. Fimmel, Y. Hong, F. Würthner and D. Kim, to be submitted.