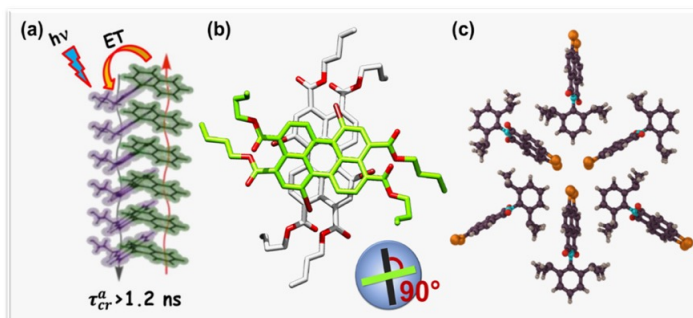


## ULTRAFAST EXCITED STATE DYNAMICS OF TWISTED AROMATICS

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**Figure.** Representative strategies adopted in our group to spatially organize organic chromophores for emergent properties.

Self-assembling of organic chromophoric systems into elegant supramolecular architectures with emergent properties has received prodigious attention in recent years.<sup>1</sup> In the first example (Figure), a naphthalene-naphthalimide donor-acceptor (D-A) dyad assembled into segregated D-A stacks in the crystalline state. The photo-induced charge separated state in the aggregate state lasts 10,000 times longer than the monomeric dyad. The femtosecond transient absorption spectra depicted the spectroscopic signature for naphthalene dimer radical cation indicating the migration of charges through the stacks.<sup>1</sup> In the second example, we report the crystalline evidence for Greek cross-

dipole ( $\alpha=90^\circ$ ) stacking of 1,7-dibromoperylene-3,4,9,10-tetracarboxylic tetrabutylester (PTE-Br2) displaying null excitonic coupling and thereby monomeric optical behavior.<sup>2</sup> In the third example, we showcase a radial assembly of 1,8-dibromonaphthalene(2,6-diisopropylphenyl)imide (NIBr2) in crystalline phase driven by hexabromine synthon.<sup>4</sup> NIBr2 exhibits ultrafast intersystem crossing<sup>3-5</sup> and solid-state room temperature phosphorescence. References: <sup>1</sup> A. R. Mallia, P. S. Salini, M. Hariharan, *J. Am. Chem. Soc.*, 137 (2015) 15604. <sup>2</sup> E. Sebastian, A. M. Philip, A. Benny, M. Hariharan, *Angew. Chem., Int. Ed.*, 57 (2018) 15696. <sup>3</sup> M. A. Niyas, R. Ramakrishnan, V. Vijay, E. Sebastian, M. Hariharan, *J. Am. Chem. Soc.*, 141 (2019) 4536. <sup>4</sup> K. Nagarajan, A. R. Mallia, K. Muraleedharan, M. Hariharan, *Chem. Sci.*, 8 (2017) 1776. <sup>5</sup> M. P. Lijina, A. Benny, R. Ramakrishnan, N. Nair, M. Hariharan, *J. Am. Chem. Soc.* 142 (2020), 17393.