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n-type doping through tethered functionality: a new paradigm for molecular design of solution-processed organic thermoelectrics BORIS RUSS, UC Berkeley, Dept of ChemE , MAXWELL J. ROBB, UCSB, Chem and Materials Depts, BHOOSHAN C. POPERE, UCSB, Dept of ChemE , ERIN E. PERRY, UCSB, Chem and Materials Depts, JEFFREY J. URBAN, LBNL, Molecular Foundry, MICHAEL L. CHABINYC, CRAIG J. HAWKER, UCSB, Chem and Materials Depts, RACHEL A. SEGALMAN, UCSB, Dept of ChemE — A scarcity of stable n-type doping mechanisms compatible with facile processing has been a major impediment to the advancement of n-type (electron transporting) organic thermoelectric materials. We recently demonstrated that trimethylammonium functionalization with hydroxide counterions, tethered to a perylene diimide core by alkyl spacers, facilitated solution-processing and resulted in extremely high carrier concentrations (10^{20}carriers/cm^3) and best-in-class thermoelectric performance in thin films. In this presentation, we report our recent findings on the underlying mechanism enabling charge carrier generation in these self-doping materials and its influence on material thermoelectric behavior. To draw these conclusions, we complement thermoelectric characterization with insights into chemical, electronic, and structural properties from XPS, optical spectroscopy, EPR, and GIWAXS experiments. Furthermore, we show that doping through tethered functionality can be extended to other n-type small molecule systems of interest, including naphthalene diimides and diketopyrrolopyrroles. Our findings help shape promising molecular design strategies for future enhancements in n-type thermoelectric performance.

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