**Designing Next-Generation Organic Semiconductors Through Phonon Engineering**

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Organic semiconductors (OSCs) are an exciting class of materials for advanced electronics. In contrast to inorganic semiconductors, OSCs can be processed in solution under benchtop conditions, enabling them to be engineered for a variety of applications, including flexible devices. However, OSCs are exhibit low charge-carrier mobilities (1-102 cm2 V-1 s-1) compared to their inorganic counterparts (103-105 cm2 V-1 s-1), limiting their utility. The origins of the reduced mobility in OSCs arise from a number of factors, but one of the most significant is electron-phonon coupling – particularly from low-frequency vibrations that are highly-excited at ambient conditions. In this work, recent efforts to generate a mode-resolved picture of electron-phonon coupling through a combined experimental and theoretical approach will be discussed, which provides insight into precisely which vibrational dynamics most strongly influence charge-carrier mobility. Such vibrations, termed ‘killer modes’ suggest that it is often only one or two mode-types that ultimately hinder charge transport in solids. Subsequently, this insight can be leveraged to mitigate detrimental phononic effects in OSCs by ‘phonon engineering’. The results of these efforts will be highlighted, and will showcase the powerful utility – as well as the supramolecular design pitfalls – that arise when subtle intermolecular forces are altered by chemical modification. Overall, this work highlights the powerful interplay between supramolecular design, electronic structure calculations, and vibrational spectroscopy for understanding electronic effects in OSCs, and suggests multiple paths forward for future work.