Understanding organic semiconductors at an atomic level: Direct simulation of the assembly of solution- and vacuum-deposited light-emitting diodes and organic photovoltaic devices.

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The optical and electronic properties of organic semiconductor thin film devices are intimately coupled to their morphology at an atomic level. Being amorphous this cannot be probed experimentally. Molecular simulations, however, allow morphologies of thin films to be examined with unrivalled spatial resolution. By directly mimicking the process by which organic semiconductor thin films are produced experimentally (solution or vacuum processing) we have generated realistic morphologies for both organic light-emitting diodes (OLEDs) and organic photovoltaics (OPVs) shedding light on factors including molecular alignment, aggregation, formation of percolation networks, interfacial effects and the presence of trapped solvent. The morphologies are highly predictive.[1-6]

For example, we showed that the higher outcoupling of OLEDs containing $Ir(ppy)_2(acac)$ as opposed to $Ir(ppy)_3$ results from kinetic-trapping during deposition; that a TAPC:C60 bulk heterojunction with 5 wt % TAPC contains extensive hole percolation pathways; and how solution deposition affects the alignment of $Ir(ppy)_2(acac)$. Emergent properties that cannot be mimicked using approximate models are also observed. The physical dimensions of the systems simulated must also correspond to real devices.

Examples demonstrating how simulations can be used to understand the connection between morphology and the optio-electronic performance of real devices will be presented, underling how simulations are becoming central to the future development of organic semiconductor devices.

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6. Sanderson, et al. (2021). **The Journal of Chemical Physics** 154 (16), 164101.