Advanced Interfacial Engineering of Edge-Contacted Graphene Devices

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The development of molecular quantum emulators is frequently hindered by polymer residues from lithography that disrupt molecular registry at 2D interfaces [1,2]. We present a modified post-fabrication purification protocol [3] that restores epitaxial graphene to UHV-equivalent purity within an edge-contacted graphene device [4]. This allows the graphene to act as a symmetry-breaking template, enforcing macroscopic long-range order in an overlayer of the organic semiconductor 2,3,6,7,10,11-hexamethoxytriphenylene (HMTP) [5]. By engineering this pristine interface, demonstrated by Low-Energy Electron Diffraction, we successfully resolve the Davydov-split excitonic manifold, Fig. 1, a hallmark of high-symmetry molecular crystals typically masked by disorder. We move beyond standard Franck-Condon models by implementing Herzberg-Teller corrections, accounting for non-Condon transitions where electronic states couple to specific vibrational modes. This approach allows for the experimental parameterization of the Holstein Hamiltonian in a functional hybrid architecture. Our findings are corroborated by Raman molecular dynamics simulations, which identify the specific vibrational modes leading to the observed excitonic phonon replicas. The results reveal a clear branching into bright and dark excitonic states, demonstrating that dark-exciton radiative channels dominate the low-energy regime. By bridging the gap between aggressive nanofabrication and high-precision surface science, this work establishes a scalable roadmap for exploring collective excitations and strong electron-phonon coupling in atomically clean molecular-2D heterostructures.

References:

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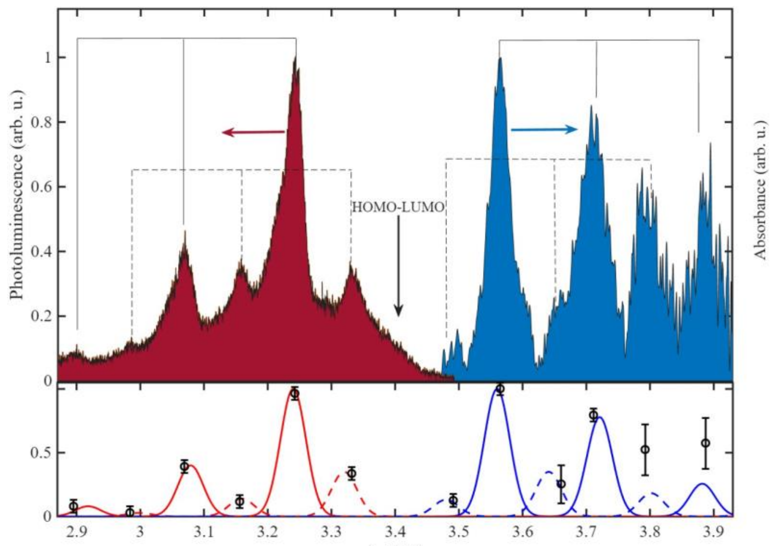

Fig. 1: Davydov-Split Excitonic Manifold in HMTP. Experimental PL and absorption spectra (top) compared with a theoretical model (bottom). The simulation incorporates a Franck-Condon mechanism with non-Condon Herzberg-Teller corrections, accurately capturing the dual sets of excitonic transitions.