Engineering the photophysics of carbon nanotube thin film devices using polaritonic microcavities

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Polaritons are hybrid light-matter states that form when matter excitations strongly interact with light in an optical cavity. These new eigenstates possess mixed properties of both matter (excitons) and light (photons), which enable photophysical and optoelectronic properties that neither possesses on their own. We present the formation and spectroscopic characterization of room-temperature exciton-polaritons in semiconducting single-walled carbon nanotube (sSWCNT) microcavities. High optical density, chirality-purified sSWCNTs are incorporated into Fabry-Perot microcavities using a lamination approach that overcomes previous challenges in fabrication that typically arise due to the inherent disorder present in randomly oriented, crossing sSWCNT dense networks. With this approach, we achieve high-quality factor microcavities that enable strong coupling as evidenced by the large Rabi splitting values extracted using a three-level Hopfield Hamiltonian. Our microcavities are used to engineer the electronic energy landscape toward delocalized excited states and long-range energy transport, which are expected to result in devices with enhanced energy and/or charge transport efficiencies. Polaritonic systems with sSWCNTs of multiple chiralities, i.e. multiple band-gap energies, are also fabricated using this lamination approach in order to investigate the impact of strong light-matter coupling on the energy diffusion dynamics. Preliminary two-dimensional white-light spectra reveal acceleration of the relaxation dynamics within the microcavity, highlighting the potential of polaritonic microcavities as a tool for tailoring and optimizing material properties toward desired device performance.