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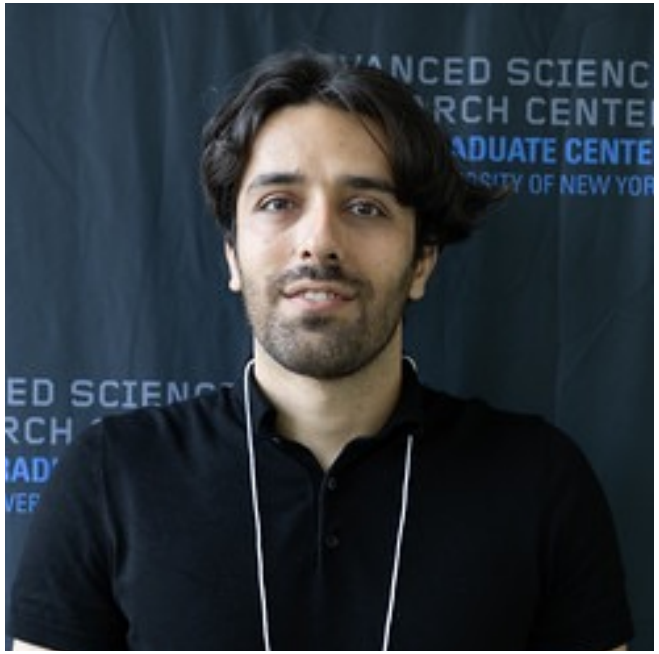
LIGHT-MATTER SEMINAR



Title: Tunable Photochemical Charge Transfer via Long-Lived Bloch Surface Wave Polaritons under Phase Matching Conditions

Abstract: The use of hybrid light-matter molecular exciton-polariton states has been proposed as a scheme to directly control and modify the efficiency and rate of photochemical phenomena. However, the efficacy of polariton-driven photochemistry remains an open question. The key limiting factor for achieving this control has been the “dark-state problem,” in which photoexcitation populates long-lived reservoir states with energies and dynamics similar to those of bare excitons. Here, we first use a sensitive ultrafast transient reflection method with momentum and spectral resolution to achieve the selective excitation of organic exciton-polaritons in open photonic cavities. We show that the energy dispersions of these systems allow us to avoid the parasitic effect of the reservoir states under phase-matching conditions, and we observe the direct population and decay of polaritons. Next, we employ different cavity designs to demonstrate conditions under which photoinduced polaritonic charge transfer can be achieved and directly visualized using momentum-resolved ultrafast spectroscopy under phase matching conditions. Key conditions for charge transfer are satisfied using Bloch surface wave polaritons, which exhibit favorable dispersion characteristics that permit the selective pumping of hybrid states with long lifetimes (100-400 fs), allowing vibrationally assisted molecular charge transfer. Using this approach, we tune the energetic driving force for charge separation, reducing it by as much as 0.5 eV compared to the bare exciton and improving the efficiency of photochemical charge transfer. These results establish that tunable and efficient polariton-driven molecular charge transfer is indeed possible under phase matching conditions and using carefully designed photonic systems.