Assessing the Material Determinants of Cavity Polariton Relaxation using Angle-Resolved Photoluminescence Excitation Spectroscopy

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Abstract : Cavity polaritons form when molecular excitons strongly couple to photons in carefully constructed optical cavities. These polaritons, which are hybrid light-matter states possessing a unique combination of photonic and excitonic properties, present the opportunity to manipulate the properties of various semiconductor materials. The systematic manipulation of materials through polariton formation could potentially improve the functionalities of many optoelectronic devices such as lasers, light-emitting diodes, photon-based quantum computers, and solar cells. However, the prospects of leveraging polariton formation for novel devices and device operation depend on more complete connections between the properties of molecular chromophores, and the hybrid light-matter states they form, which remains an outstanding scientific goal. Specifically, for most optoelectronic applications, it is paramount to understand how polariton formation affects the spectra of light absorbed by molecules coupled strongly to cavity photons. An essential feature of a polariton state is its dispersive energy, which occurs due to the enhanced spatial delocalization of the polaritons relative to bare molecules. To leverage the spatial delocalization of cavity polaritons, angle-resolved photoluminescence excitation spectroscopy was employed in characterizing light emission from the polaritonic states. Using lasers of appropriate energies, the polariton branches were resonantly excited to understand how molecular light absorption changes under different strong light-matter coupling conditions. Since an excited state has a finite lifetime, the photon absorbed by the polariton decays non-radiatively into lower-lying molecular states, from which radiative relaxation to the ground state occurs. The resulting fluorescence is collected across several angles of excitation incidence. By modeling the behavior of the light emission observed from the lower-lying molecular state and combining this result with the output of angle-resolved transmission measurements, inferences are drawn about how the behavior of molecules changes when they form polaritons. These results show how the intrinsic properties of molecules, such as the excitonic lifetime, affect the rate at which the polaritonic states relax. While it is true that the lifetime of the photon mediates the rate of relaxation in a cavity, the results from this study provide evidence that the lifetime of the molecular exciton also limits the rate of polariton relaxation.

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