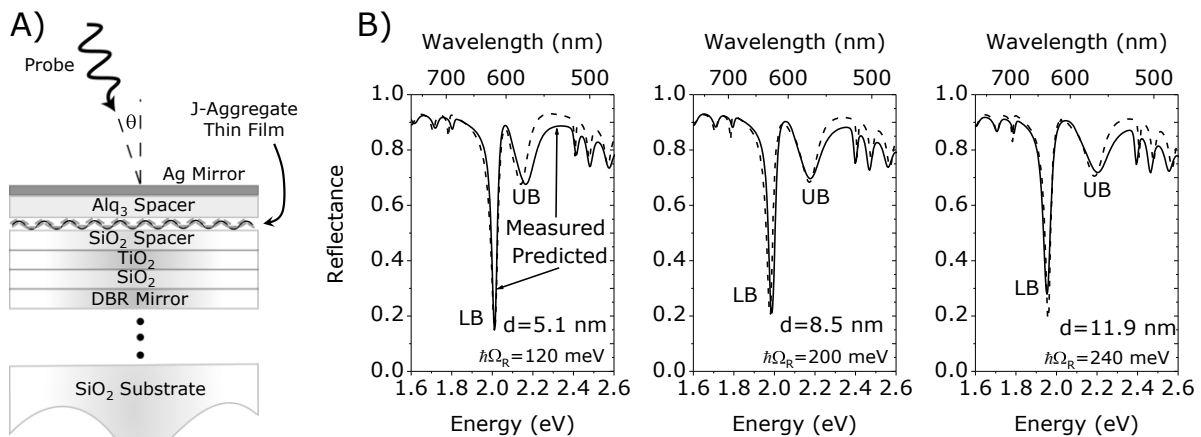


Predicting the Linear Optical Response of J-Aggregate Microcavity Exciton-Polariton Devices

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We use the transfer matrix numerical formalism to accurately predict the linear optical properties of strongly-coupled microcavity exciton-polariton devices containing thin films of J-aggregated cyanine dyes. Thin films of cyanine dye J-aggregates enable the observation of strong coupling between light and matter at room temperature due to their high absorption constant and narrow linewidth, which allowed for the first demonstration of exciton-polariton electroluminescence at room temperature [1-4]. Our model uses the spectrally-resolved complex index of refraction of the TDBC (5,6-dichloro-2-[3-[5,6-dichloro-1-ethyl-3-(3-sulfopropyl)-2(3H)-benzimidazolide]-1-propenyl]-1-ethyl-3-(3-sulfopropyl) benzimidazolium hydroxide, inner salt, sodium salt) J-aggregate films [5]. The index of refraction is calculated by model-free quasi-Kramers-Kronig regression performed on the reflectance measurements of neat J-aggregate films on glass, enabling us to numerically engineer the properties of the complete microcavity devices (see Figure A) prior to fabrication. We demonstrate robustness of this numerical method by matching the experimentally obtained angular dispersion of light reflected from the strongly-coupled microcavity to the predictions of our model. Both numerically and experimentally we demonstrate that the Rabi splitting of exciton-polariton modes of strongly-coupled microcavities can be finely tuned by modifying the J-aggregate film thickness (see Figure B).



(A) J-aggregate microcavity exciton-polariton device structure. (B) Comparison of measured and predicted reflectance spectra for device shown. Upper and lower branch (UB and LB) polaritons are indicated.

Finally, we apply the model to estimate the linewidth of the lower branch exciton-polaritons in J-aggregate-based structures to be 8.4 meV. This is a key parameter in theoretical modeling of exciton-polariton dynamics and more than a factor of two smaller than linewidths assumed in previous studies [6-8].

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