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Quantum Dynamics and Emission Power Spectra of Molecules in Complex Dielectric Environments: Studies based on Macroscopic Quantum Electrodynamics

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Abstract

Based on Macroscopic quantum electrodynamics (MQED), we developed theories of single-molecule emission power spectra and multichromophoric excitation energy transfer. These theories serve as powerful tools for investgating effects of light-matter interaction in complex dielectric environments. In addition, we also connected the MQED theory with the dissipative Tavis-Cummings model and provided a simple standard that enables one to examine the validity of the (dissipative) Tavis-Cummings model in different photonic environments.

Macroscopic quantum electrodynamics

$$\hat{H} = \hat{H}_{\mathrm{M}} + \hat{H}_{\mathrm{P}} + \hat{H}_{\mathrm{I}}$$

$\hat{H}_{\mathrm{M}} \longrightarrow$ Molecules

$\hat{H}_{\mathrm{P}} = \int \mathrm{d}^3\mathbf{r} \int_0^\infty \mathrm{d}\omega\, \hbar\omega \hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}(\mathbf{r},\omega) \longrightarrow$ Bosonic Vector Fields

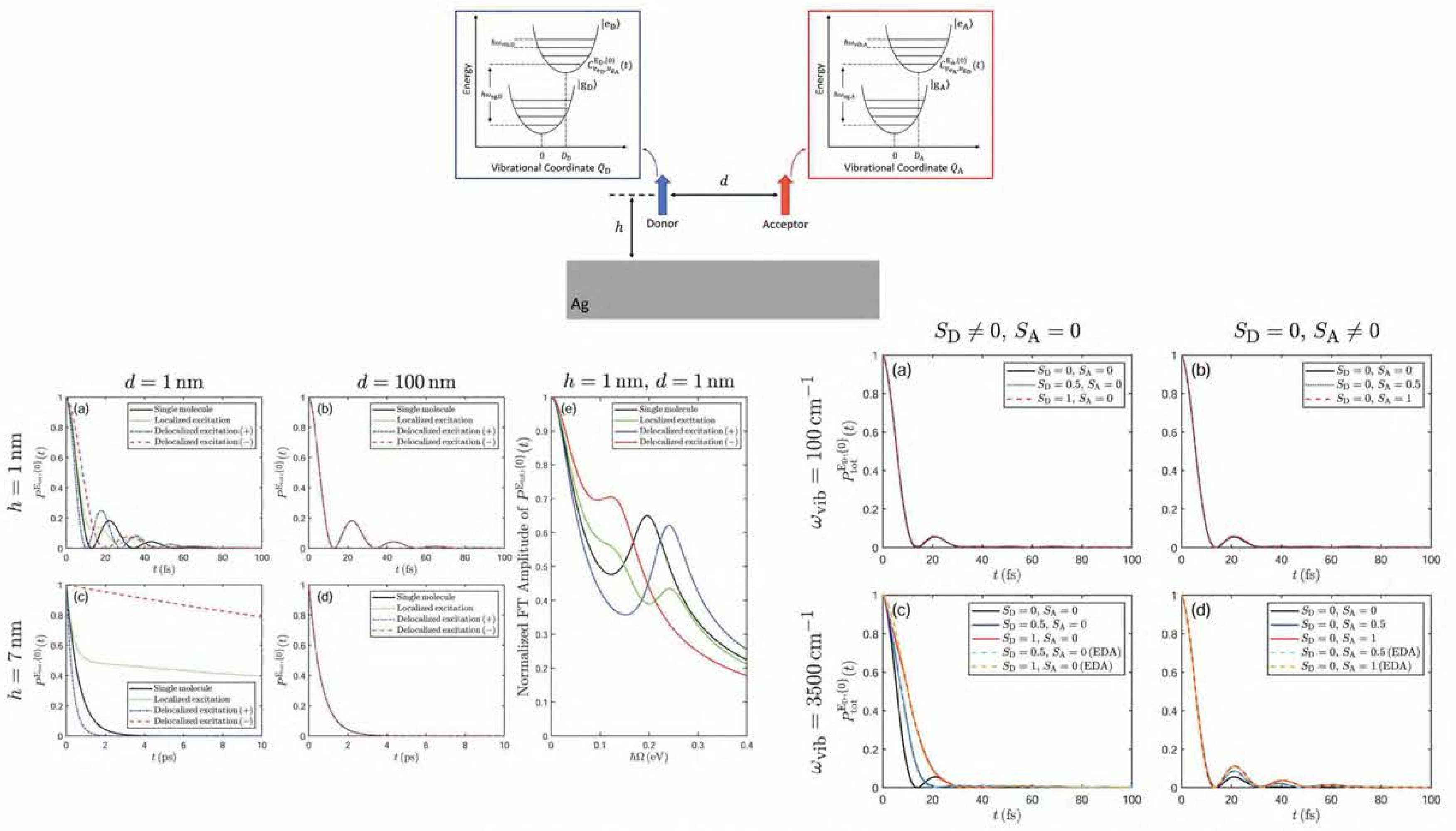
$\hat{H}_{\mathrm{I}} = -\sum_{\zeta=1}^{N}\left[\hat{\sigma}_\zeta^{(+)} + \hat{\sigma}_\zeta^{(-)}\right]\mu_\zeta \mathbf{n}_\zeta \cdot \boxed{\hat{\mathbf{E}}(\mathbf{r}_\zeta)} \longrightarrow$ Light-Matter Couplings

$$\boxed{\hat{\mathbf{E}}(\mathbf{r}_\zeta) \equiv i\sqrt{\frac{\hbar}{\pi\varepsilon_0}} \int_0^\infty \mathrm{d}\omega \int \mathrm{d}^3\mathbf{r}\, \frac{\omega^2}{c^2}\sqrt{\mathrm{Im}\left[\varepsilon_{\mathrm{r}}(\mathbf{r},\omega)\right]}\, \boxed{\overline{\overline{\mathbf{G}}}(\mathbf{r}_\zeta,\mathbf{r},\omega)} \cdot \hat{\mathbf{f}}(\mathbf{r},\omega) + \mathrm{H.c.}}$$

$$\left[\frac{\omega^2}{c^2}\varepsilon_{\mathrm{r}}(\mathbf{r}_\zeta,\omega) - \nabla\times\nabla\times\right]\boxed{\overline{\overline{\mathbf{G}}}(\mathbf{r}_\zeta,\mathbf{r},\omega)} = -\overline{\overline{\mathbf{I}}}_3\delta(\mathbf{r}_\zeta - \mathbf{r})$$

Macroscopic Maxwell's Equations

Multichromophoric excitation energy transfer



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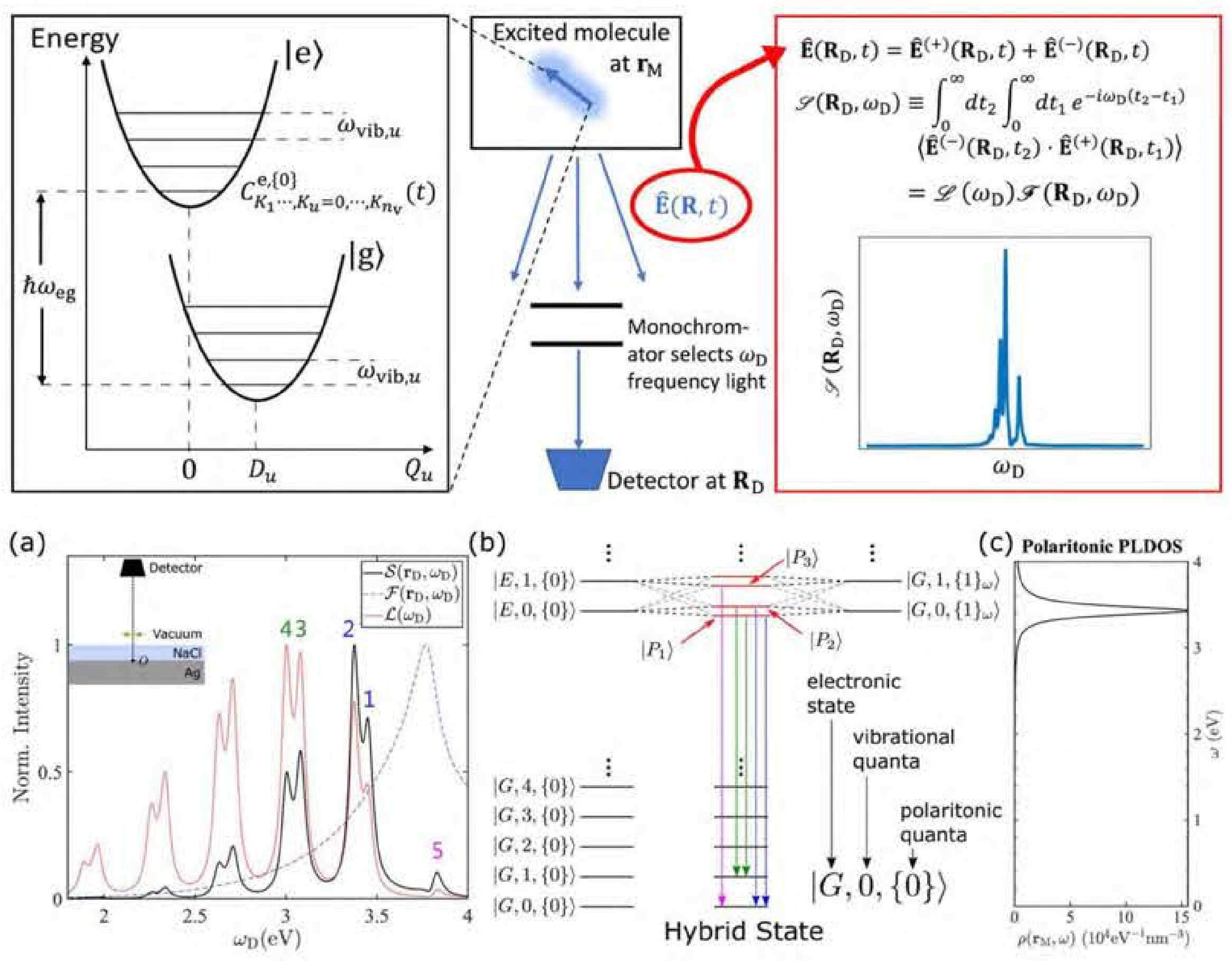
Single-molecule emission power spectra



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Revision of Tavis-Cummings Model

$$\mathcal{J}(\mathbf{r}_\zeta,\mathbf{r}_{\zeta'},\omega) \equiv \frac{\mu^2\omega^2}{\pi\hbar\varepsilon_0 c^2}\mathbf{n}_\zeta \cdot \mathrm{Im}\left[\overline{\overline{\mathbf{G}}}(\mathbf{r}_\zeta,\mathbf{r}_{\zeta'},\omega)\right]\cdot\mathbf{n}_{\zeta'} \sim \frac{g^2}{\pi}\frac{\kappa_{\mathrm{ph}}/2}{(\omega-\omega_{\mathrm{ph}})^2 + (\kappa_{\mathrm{ph}}/2)^2}$$

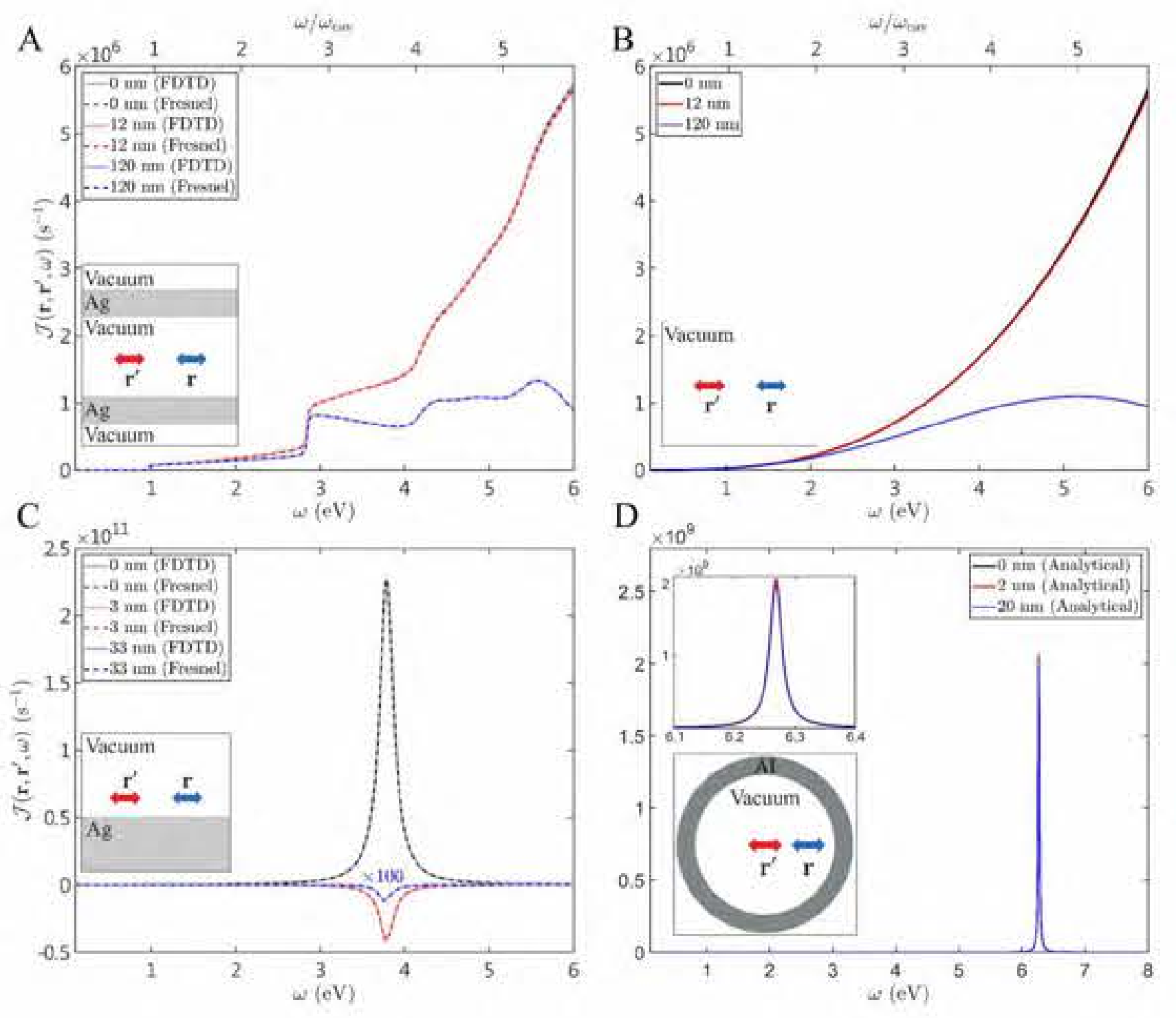
$$\frac{\mathrm{d}}{\mathrm{d}t}\hat{\rho}_{\mathrm{eff}} = -\frac{i}{\hbar}\left[\boxed{\hat{H}_{\mathrm{TC}}}\,\hat{\rho}_{\mathrm{eff}}\right] + \kappa_{\mathrm{ph}}\boxed{L_{\hat{a}}}\left[\hat{\rho}_{\mathrm{eff}}\right]$$

Effective Tavis-Cummings Model:

$$\hat{H}_{\mathrm{TC}} \equiv \hat{H}_{\mathrm{M}} + \hbar\omega_{\mathrm{ph}}\hat{a}^\dagger\hat{a} + \sum_\zeta \hbar g\left[\hat{\sigma}_\zeta^{(+)}\hat{a} + \hat{\sigma}_\zeta^{(-)}\hat{a}^\dagger\right]$$

Lindblad Dissipator (Loss):

$$L_{\hat{a}}\left[\hat{\rho}_{\mathrm{eff}}\right] \equiv \hat{a}\hat{\rho}_{\mathrm{eff}}\hat{a}^\dagger - \frac{1}{2}\left\{\hat{a}^\dagger\hat{a}, \hat{\rho}_{\mathrm{eff}}\right\}$$



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研究生活及心得

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