Spontaneous emission from quantum dots beyond Fermi's golden rule

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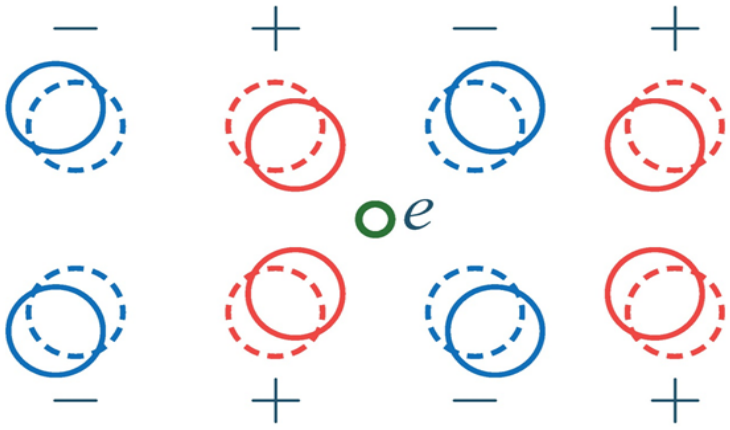
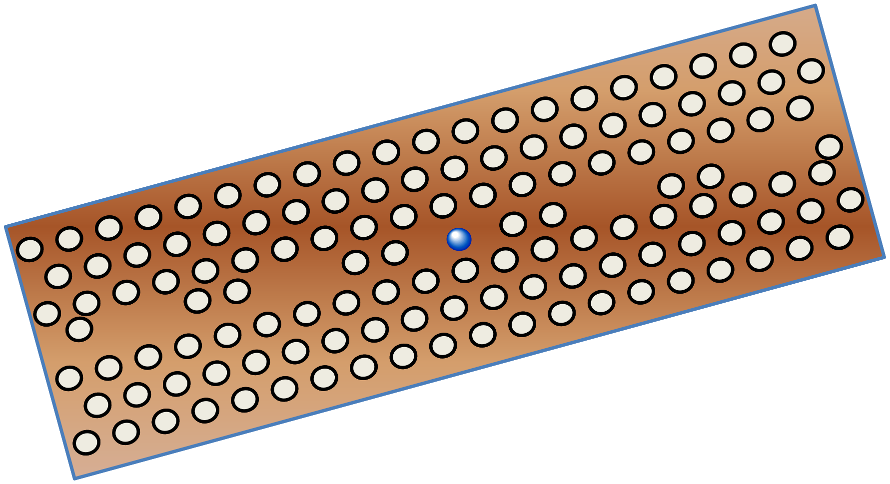
Thanks:

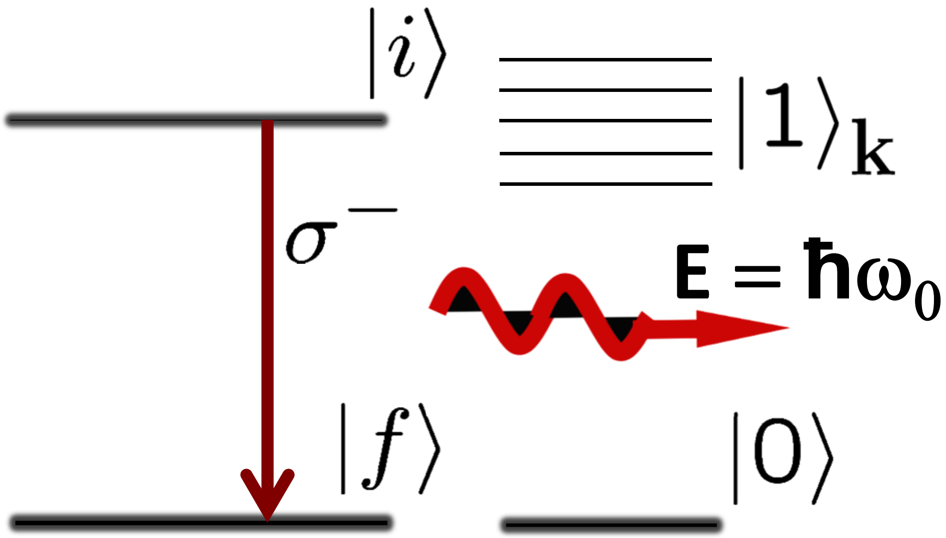
Kaushik Roy-Choudhury, NSERC($), organizers





Fermi’s golden rule for spon. emission

$$\boxed{\Gamma_{fi} = \frac{2\pi}{\hbar^2}\rho(\omega_{\mathbf{k}})\left|\langle f, 1_{\mathbf{k}}|H_{\text{Int}}|0_{\mathbf{k}}\rangle\right|^2}$$

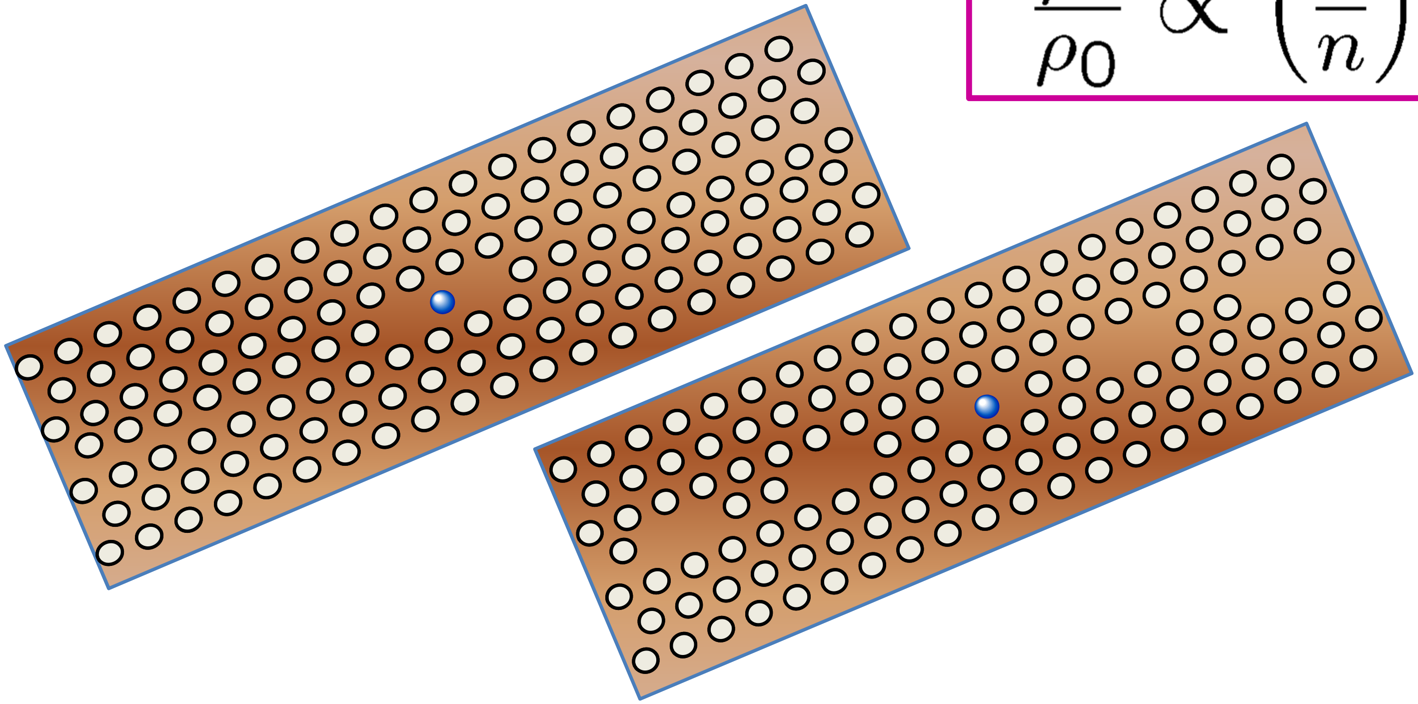


LDOS: $\rho(\mathbf{r}_0, \omega_0) \propto \text{Im}[\mathbf{G}(\mathbf{r}_0, \mathbf{r}_0, \omega_0)]$

Purcell effect

- LDOS can be tuned by having QDs coupled to resonant cavity structures, e.g., in photonic crystals, which can enhance the SE rate

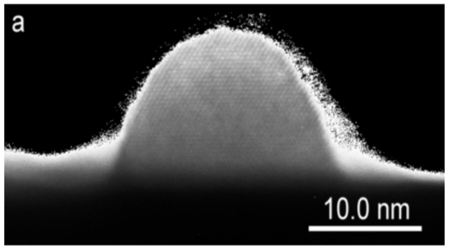
$$\boxed{\frac{\rho_c}{\rho_0} \propto \left(\frac{\lambda}{n}\right)^3 \frac{Q}{V}}$$



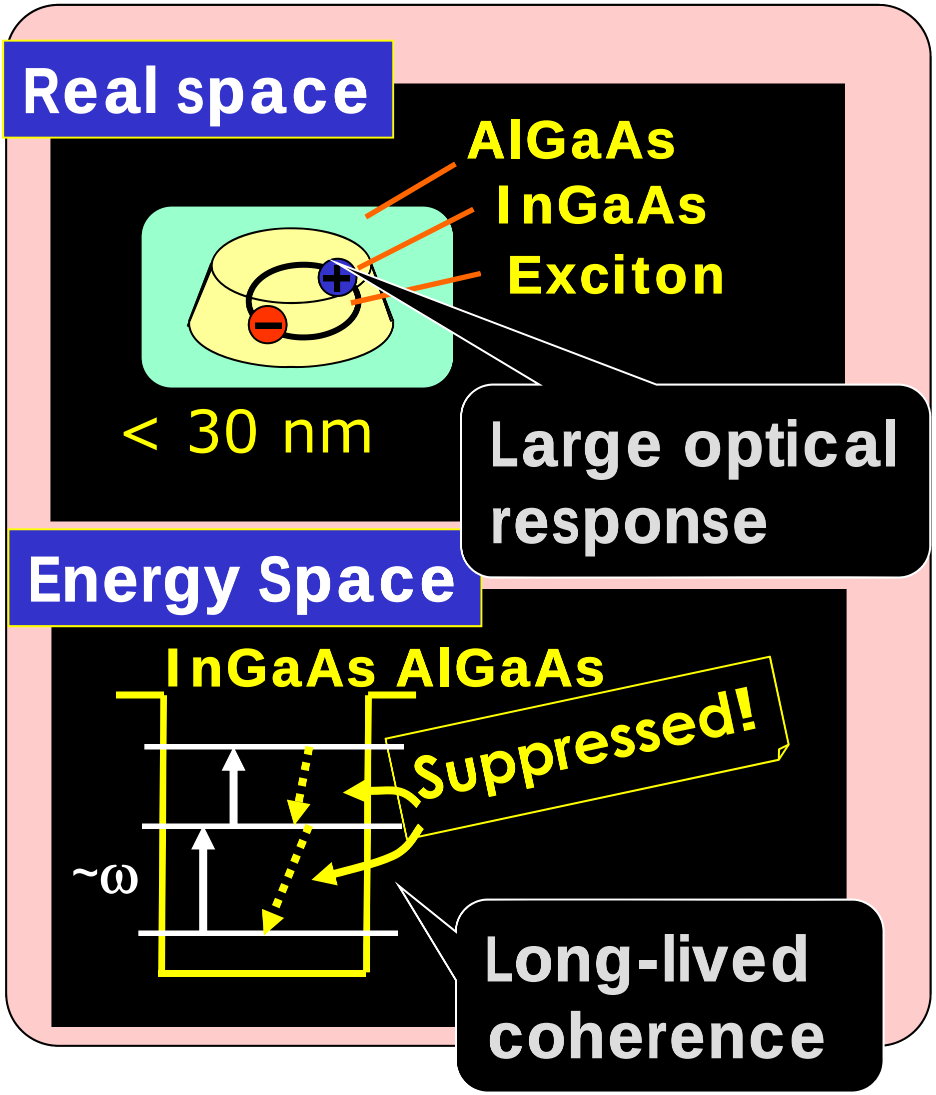
E. M. Purcell, Phys. Rev. 69, 681 (1946).

Semiconductor quantum dots (QDs)

InAs dot, NRC, Canada



- 3D confinement
- Discrete density-of-states
- Usually inside solid crystal



Real space

< 30 nm

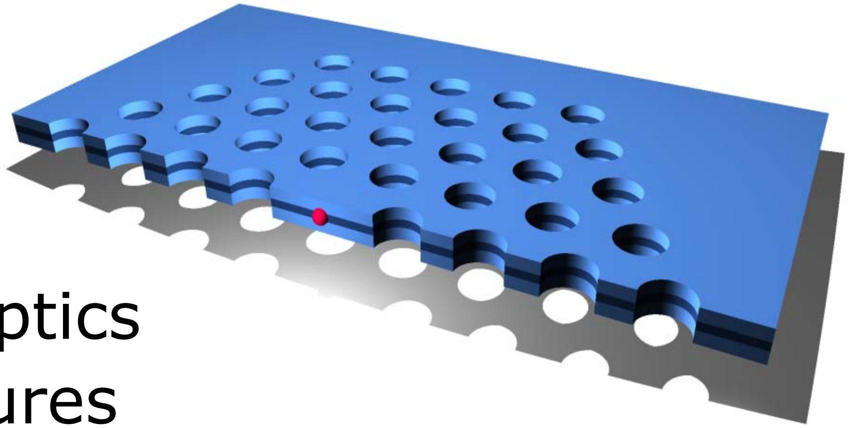
Large optical response

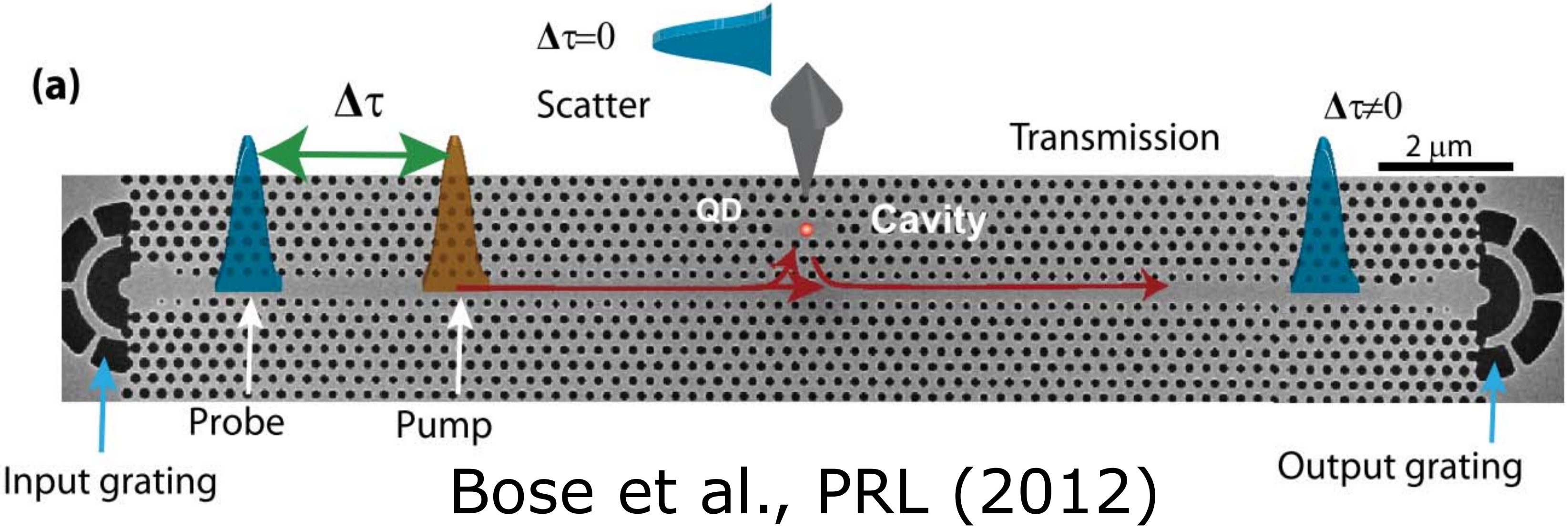
Energy Space

Long-lived coherence

Motivation for QD quantum optics

- Integration, tunable emission, large dipoles
- On-chip QIP
- Fundamental quantum optics in solid state nanostructures





Bose et al., PRL (2012)

Outline

- Basic theory of SE in a photon bath
- Coupling in an acoustic phonon bath – polaron transform approach
- Mean field limits and connection to previous works – why Fermi's golden rule breaks down?
- Examples for a cavity and a coupled cavity waveguide

Light-Matter Hamiltonian

$$\begin{aligned} H = \int d\mathbf{r} \int_0^\infty d\omega\, \omega\, \hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\hat{\mathbf{f}}(\mathbf{r},\omega) + \omega_x \hat{\sigma}^+ \hat{\sigma}^- \\ - \left[\hat{\sigma}^+ \int_0^\infty d\omega\, \mathbf{d}\cdot\hat{\mathbf{E}}(\mathbf{r}_d,\omega) + \mathrm{H.c.}\right] \end{aligned}$$

With: $\hat{\mathbf{E}}(\mathbf{r},\omega) \propto \int d\mathbf{r}' \mathbf{G}(\mathbf{r},\mathbf{r}';\omega)\sqrt{\mathrm{Im}[\varepsilon(\mathbf{r}',\omega)]}\hat{\mathbf{f}}(\mathbf{r}',\omega)$

Applies to any inhomogeneous and lossy structure, including metal nanoparticles.

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e.g., S. Scheel, L. Knoll, D.-G. Welsch, PRA 60 4094 (1999)

Master Equation in Interaction Picture

Interaction picture, 2nd-order Born approximation:

$$\frac{\partial\tilde{\rho}(t)}{\partial t} = -\int_0^t d\tau\,\mathrm{Tr}_\mathrm{R}\{[\tilde{H}_\mathrm{I}(t),[\tilde{H}_\mathrm{I}(t-\tau),\tilde{\rho}(t)\rho_\mathrm{R}]]\}$$

$$\tilde{H}_\mathrm{I}(t) = -[\hat{\sigma}^+\int_0^\infty d\omega\,\hat{\mathbf{E}}(\mathbf{r}_d,\omega)e^{(\omega_x-\omega)t} + \mathrm{H.c.}]$$

Bath approximation, gives Lindblad scattering term

$$\left.\frac{\partial\rho}{\partial t}\right|_\mathrm{inc} = \frac{\gamma(t)}{2}(2\hat{\sigma}^-\rho\hat{\sigma}^+ - \hat{\sigma}^+\hat{\sigma}^-\rho - \rho\hat{\sigma}^+\hat{\sigma}^-)$$

Spontaneous emission rate in a photon bath

$$\gamma(t) = \mathrm{Re}(\textstyle\int_0^t J_{\mathrm{ph}}(\tau))$$

Photon correlation function

$$J_{\mathrm{ph}}(\tau) = \int_0^\infty d\omega \frac{\mathbf{d} \cdot \mathrm{Im}[\mathbf{G}(\mathbf{r}_d, \mathbf{r}_d; \omega)] \cdot \mathbf{d}}{4\pi\hbar\varepsilon_0} e^{i(\omega_x - \omega)\tau}$$

“Golden rule” obtained in Markov limit

$$\gamma \equiv \gamma(t \to \infty) \propto \mathrm{LDOS}(\omega_x)$$

For better or worse – QDs are not atoms!

Besombes et al, PRB, 2001

Acoustic phonon broadening mechanism in single quantum dot emission

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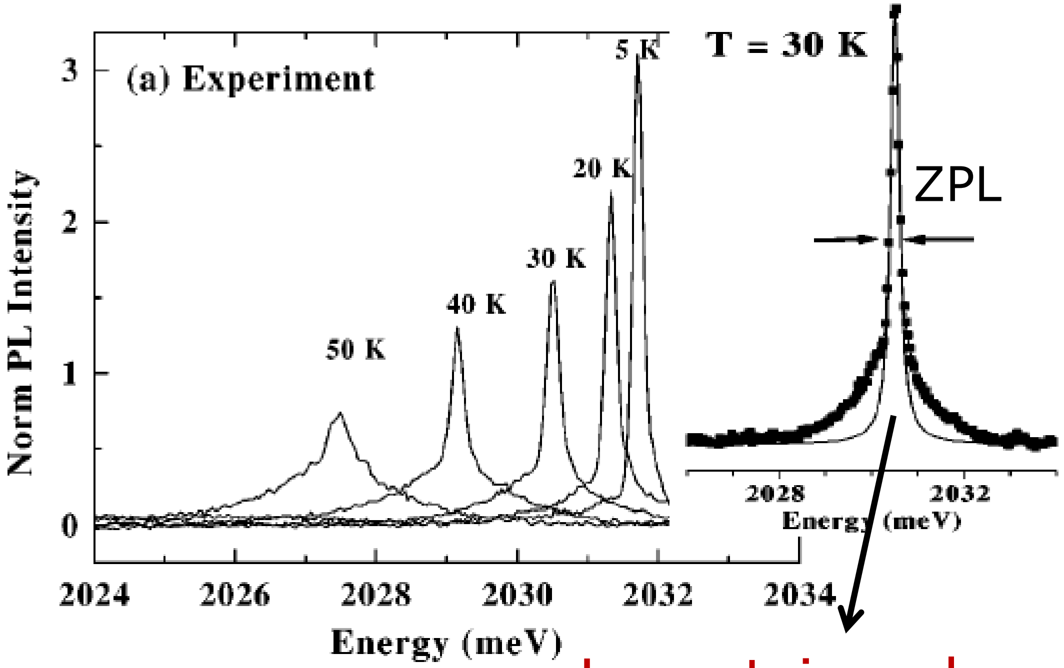
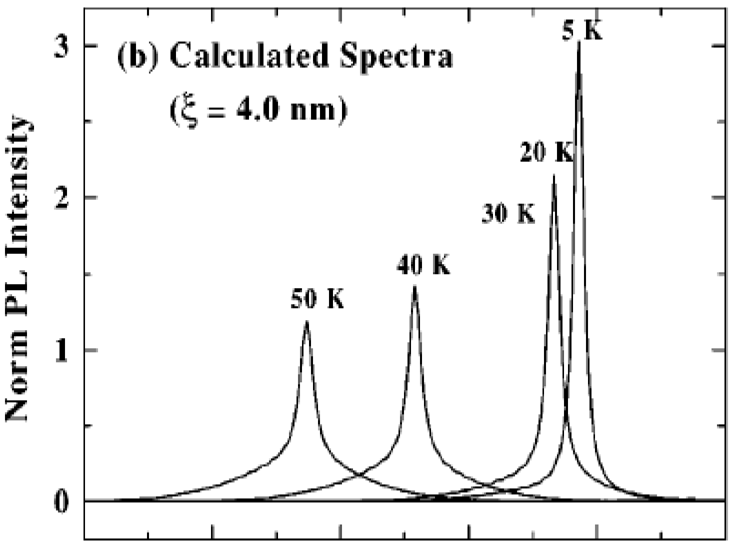


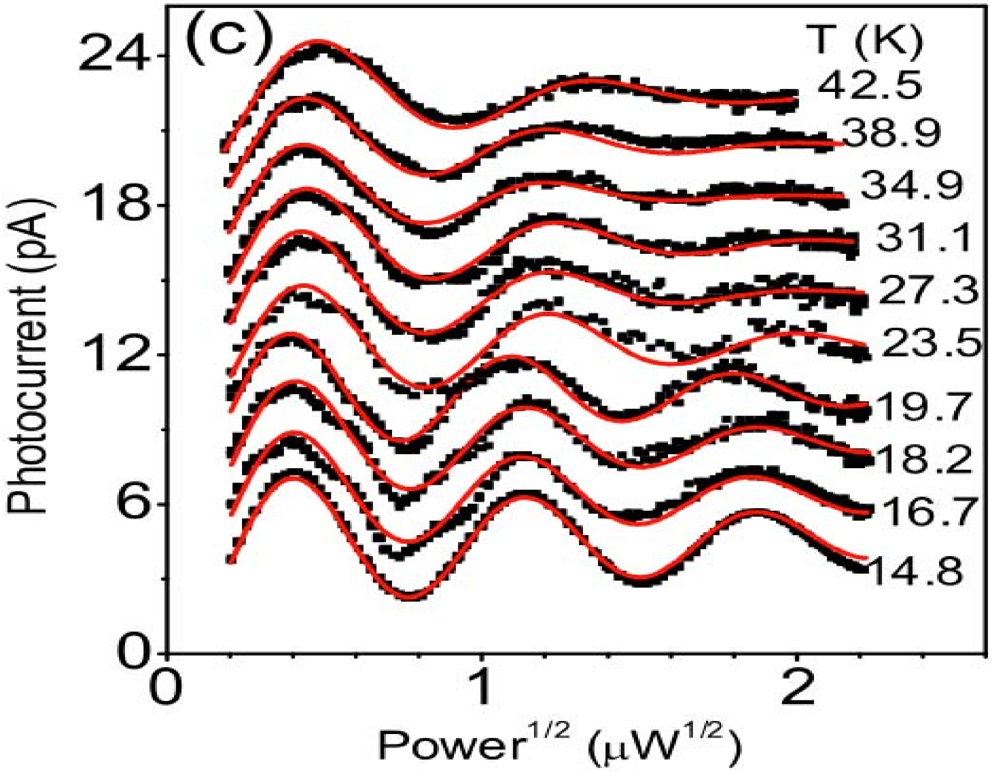
Lorentzian plus phonon sidebands



Independent boson model (IBM)

Excitation-Induced Dephasing (EID)

"Damping of exciton Rabi rotations by acoustic phonons …"



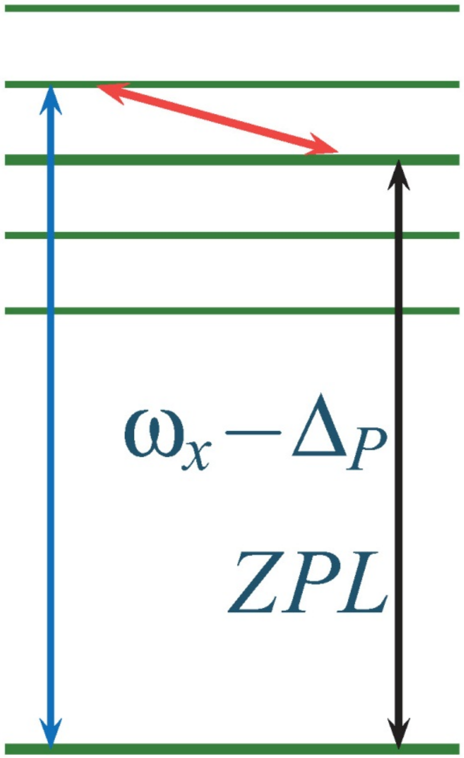
Dephasing Rate:

$$\boxed{\begin{array}{l}\Gamma \propto \Omega^2 \\ \Gamma \propto T\end{array}}$$

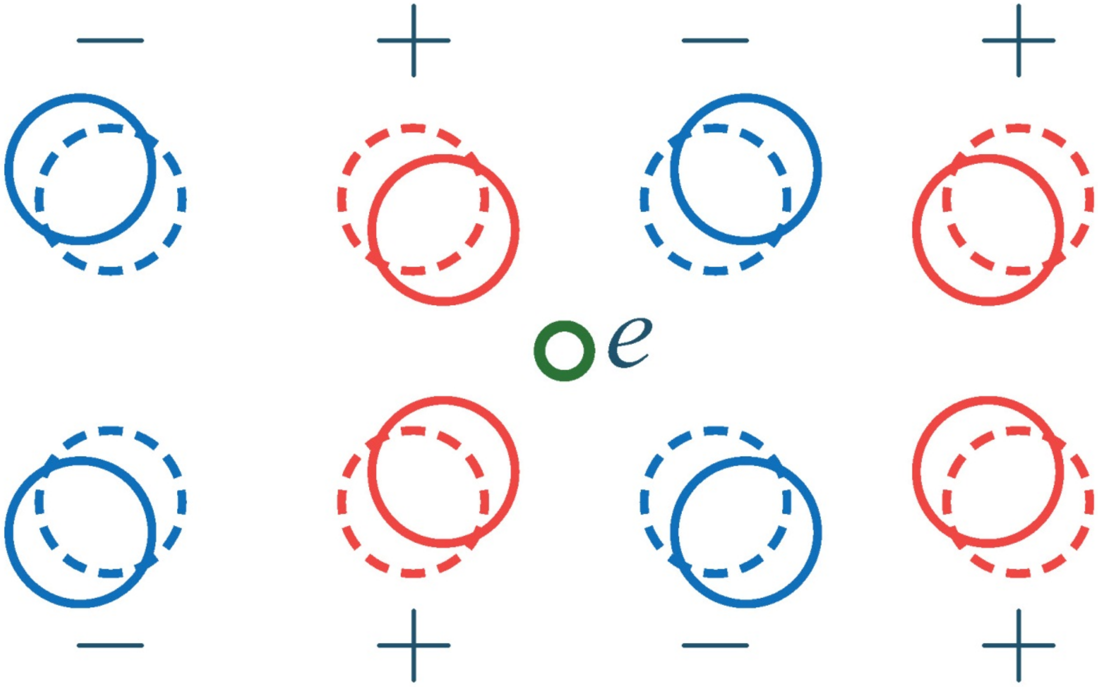
Ramsey et al, PRL 104, 017402 (2010)

Theory: see also Forstner et al, PRL (2003)

Electron+Phonons: “Polarons”



electrons in an ionic lattice:

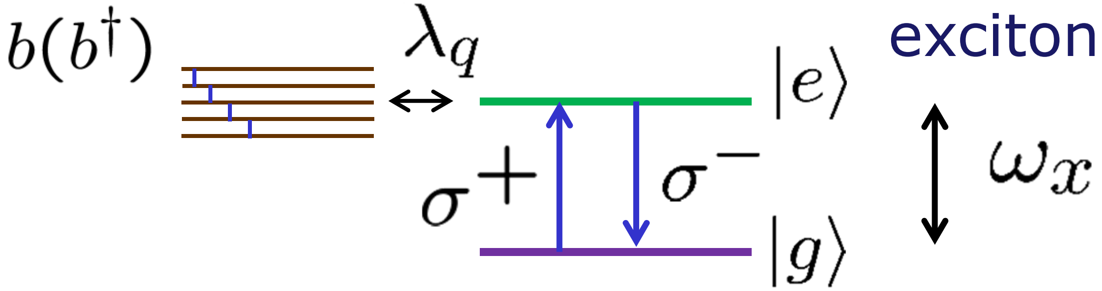


deformation (self) potential arises that acts back on electron

Independent Boson Model (IBM)

$$H_{\mathrm{IBM}} = \omega_x \sigma^+ \sigma^- + \sum_q \omega_q b_q^\dagger b_q + \sigma^+ \sigma^- \sum_q \lambda_q (b_q + b_q^\dagger)$$

LA phonon bath



- *Exactly solvable model* for <u>linear</u> polarization decay:

$$P(t) \propto e^{-i(\omega_x - \Delta_P)t} e^{[\phi(t) - \phi(0)]}$$

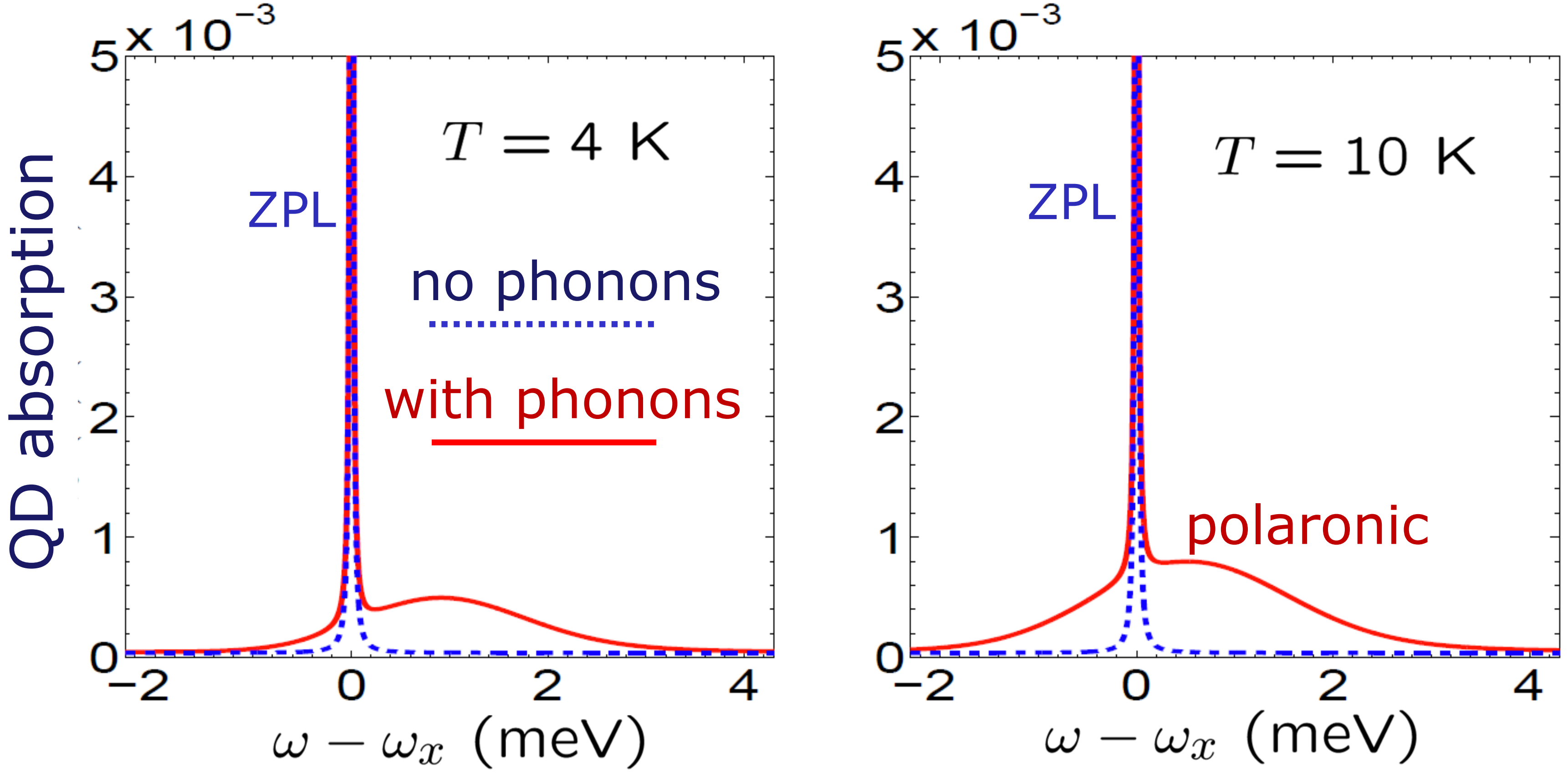
$$\phi(t) = \sum_k \frac{\lambda_q}{\omega_q} \left[\underbrace{N_q e^{i\omega_q t}}_{\text{emission}} + \underbrace{(N_q + 1)}_{\text{absorption}} e^{-i\omega_q t} \right]$$

see Axt et al, Knorr et al, Mahan's "Many Particle Physics" book

Phonon Spectral Function/QD Absorption

$$J(\omega) = \alpha_p\,\omega^3 \exp\left[-\frac{\omega^2}{2\omega_b^2}\right] \qquad \omega_b = 1\,\mathrm{meV} \qquad \alpha_p/(2\pi)^2 = 0.06\,\mathrm{ps}^2$$

InAs QDs*



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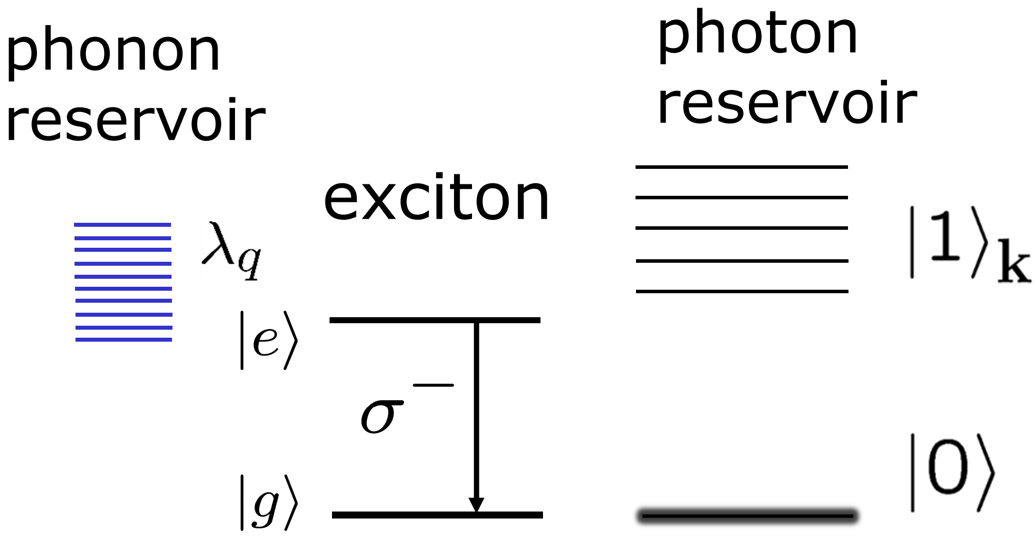
* See SH et al., PRB 83, 165313 (2011)

Coupling Photon and Phonon Baths

$$\begin{aligned} H = &\int d\mathbf{r} \int_0^\infty d\omega\, \omega\, \hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\hat{\mathbf{f}}(\mathbf{r},\omega) + \omega_x \hat{\sigma}^+\hat{\sigma}^- \\ &- \left[\hat{\sigma}^+ \int_0^\infty d\omega\, \mathbf{d}\cdot\hat{\mathbf{E}}(\mathbf{r}_d,\omega) + \text{H.c.}\right] \\ &+ \sigma^+\sigma^- \sum_q \lambda_q (b_q + b_q^\dagger) + \sum_q \omega_q b_q^\dagger b_q \end{aligned}$$



Master Equation with Photons and Phonons

- Polaron transform

$H' \rightarrow e^{P} H e^{-P}$ with $P = \sigma^{+}\sigma^{-}\sum_q \frac{\lambda_q}{\omega_q}(b_q^{\dagger} - b_q)$

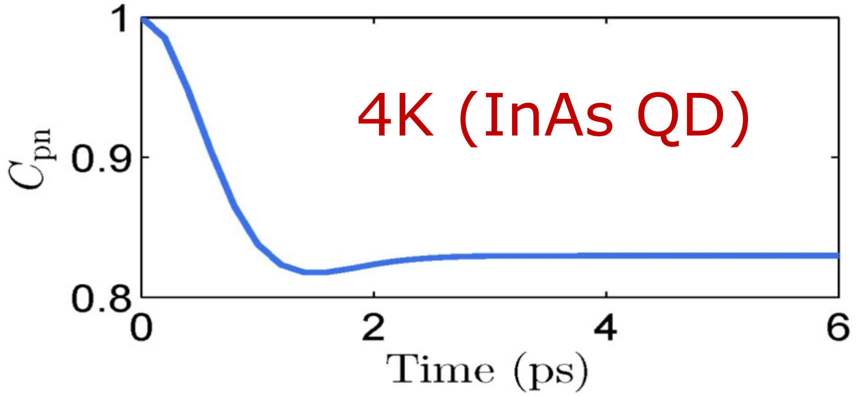
- Phonon correlation function: $C_{\text{pn}}(t) = e^{[\phi(t)-\phi(0)]}$
- Phonon-modified SE rate:

$$\begin{aligned}\tilde{\gamma}(t) &= \text{Re}\Big[\int_0^t d\tau\, C_{\text{pn}}(\tau) \int_0^{\infty} d\omega\, J_{\text{ph}}(\omega) e^{i(\omega_x-\omega)\tau}\Big] \\ &= \text{Re}\Big[\int_0^t d\tau\, C_{\text{pn}}(\tau) J_{\text{ph}}(\tau)\Big]\end{aligned}$$

$\tilde{\gamma} \not\propto \text{LDOS}(\omega_x)$ ⇨ Breakdown of golden rule

Mean Field Limits (both golden rule)

$$\tilde{\gamma}(t) = \mathrm{Re}[\int_0^t d\tau\, C_{\mathrm{pn}}(\tau) J_{\mathrm{ph}}(\tau)]$$



Delta function photon correlation time (free space)

$$\tilde{\gamma} = \mathrm{Re}(\int_0^\infty J_{\mathrm{ph}}(\tau)) \rightarrow \gamma(t)$$

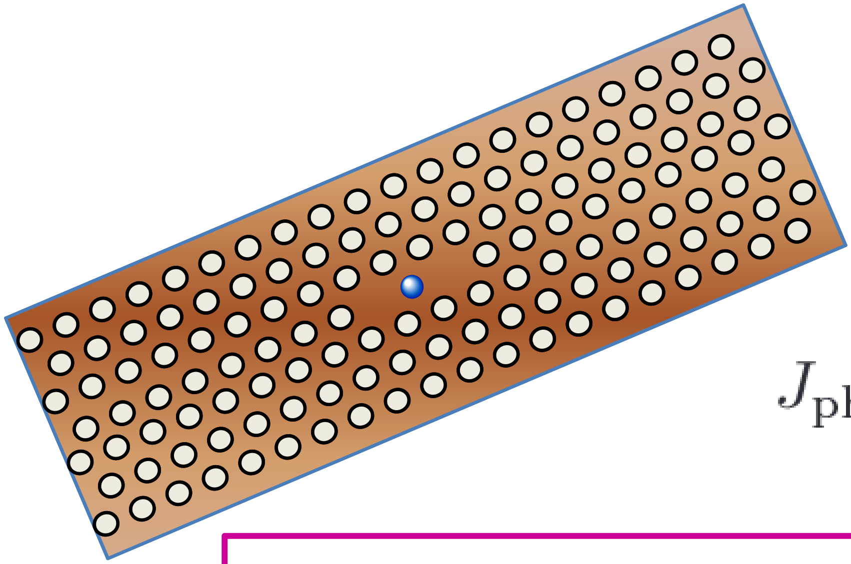
McCutcheon, Nazir, PRL. 2013; P. Kaer et al. PRB, 2012.

Long photon bath correlation time (high Q cavity)

$$\tilde{\gamma}(t) = e^{-\phi(0)}\mathrm{Re}(\int_0^t e^{\phi(\tau)} J_{\mathrm{ph}}(\tau)) \rightarrow \langle B\rangle^2 \gamma(t)$$

Roy and John, PRA, 2010

Example 1: Simple Lorentzian Cavity



Photon bath function:

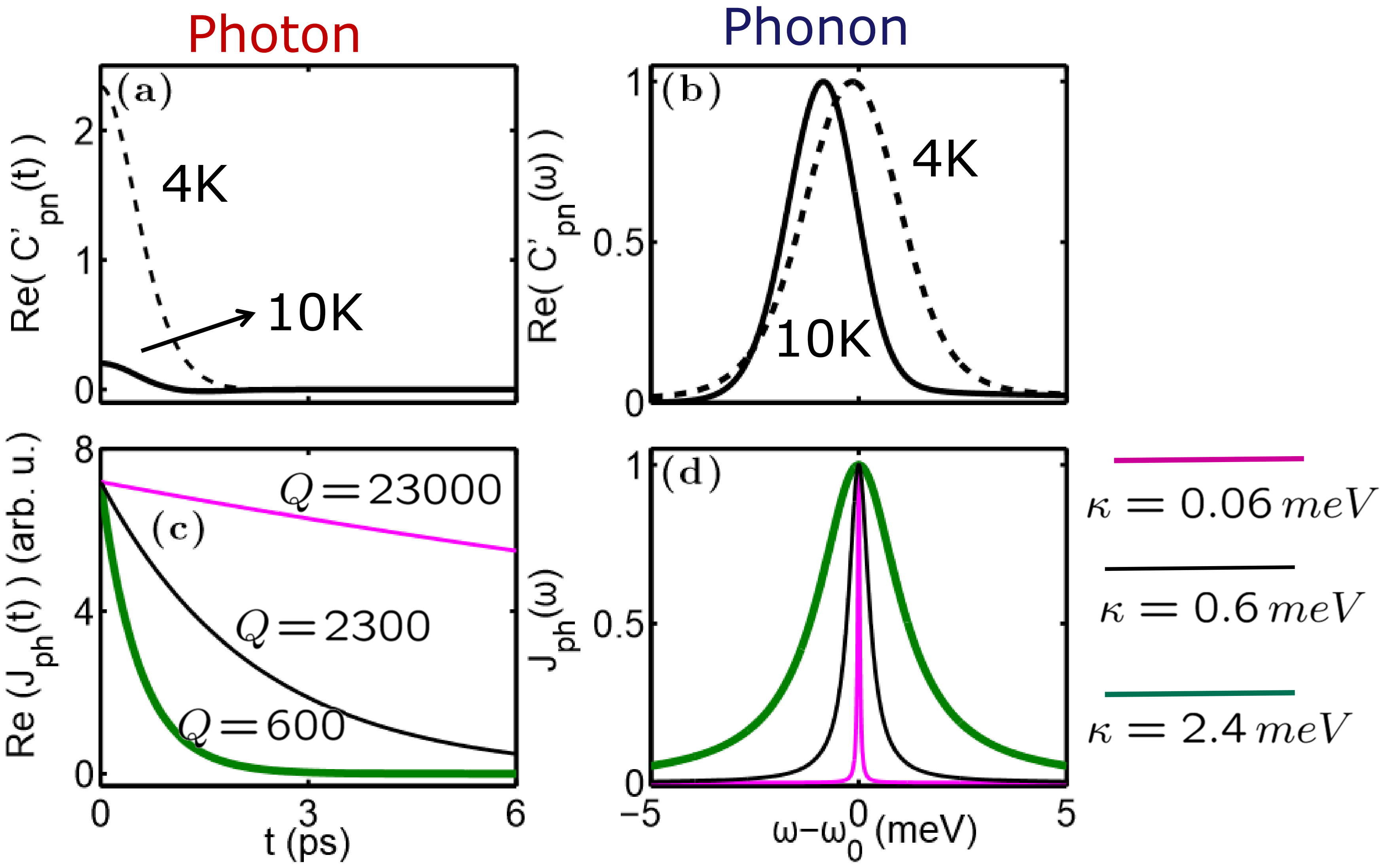
$$J_{\mathrm{ph}}(\omega) = g^2 \frac{1}{\pi} \frac{\frac{\kappa}{2}}{(\omega - \omega_c)^2 + (\frac{\kappa}{2})^2}$$

$$\boxed{\tilde{\gamma} = 2g^2 \langle B \rangle^2 \, \mathrm{Re} \left[\int_0^\infty e^{\phi(\tau)} e^{-i\Delta_{cx}\tau - \kappa\tau/2} d\tau \right]}$$

$$\Rightarrow \quad \mathrm{PF}_{\mathrm{QD}} = \left[\frac{3}{4\pi^2} \left(\frac{\lambda_0}{n_b} \right)^3 \frac{Q}{V_{\mathrm{eff}}} \left(\frac{\frac{\kappa^2}{4}}{\Delta_{cx}^2 + \frac{\kappa^2}{4}} \right) \right] \chi(T)$$

Phonon-modified Purcell factor

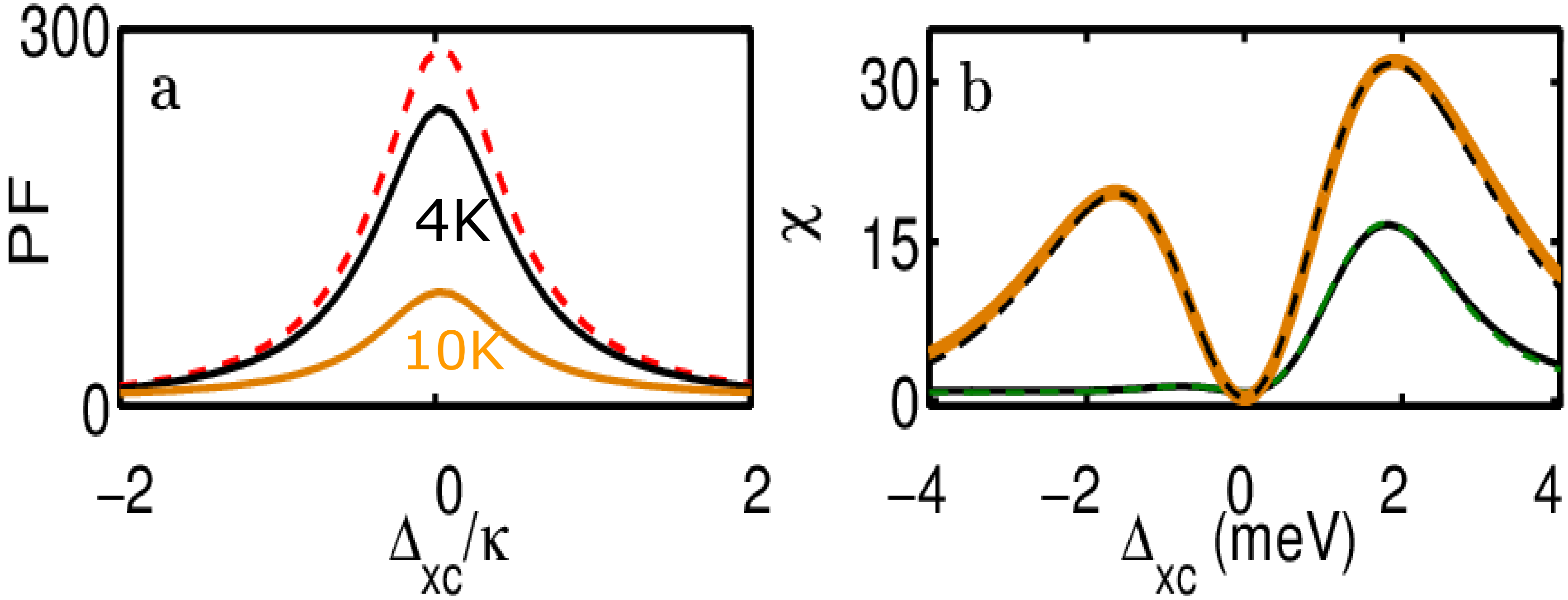
Three Example Cavity Bath Functions



$$C'_{\text{pn}}(t) = e^{[\phi(t)]} - 1$$

Phonon-Modified SE: High Q

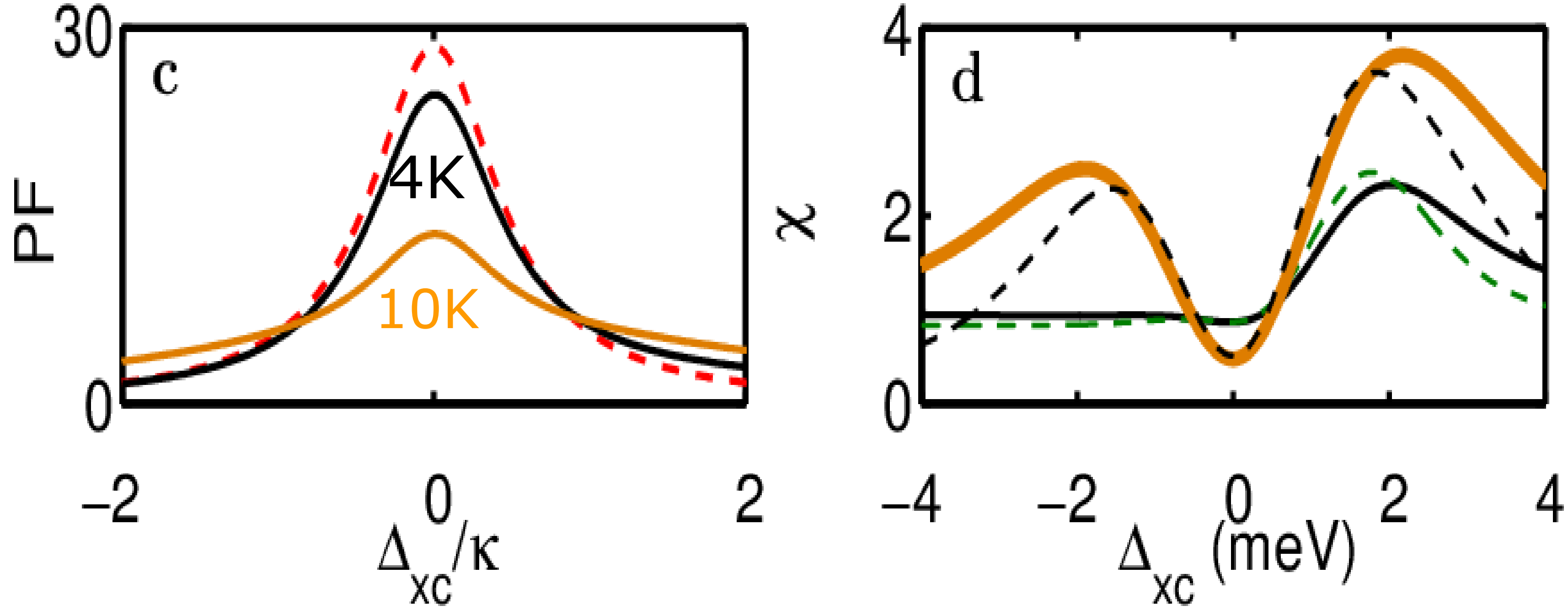
$Q = 23000 \quad \kappa = 0.06\,meV$



Dashed lines on right are results of previous cavity-QED polaron MEs, where mean field limit is used for SE decay (Wilson-Rae, Imamoglu PRB,2002; Roy, SH, PRL, 2011)

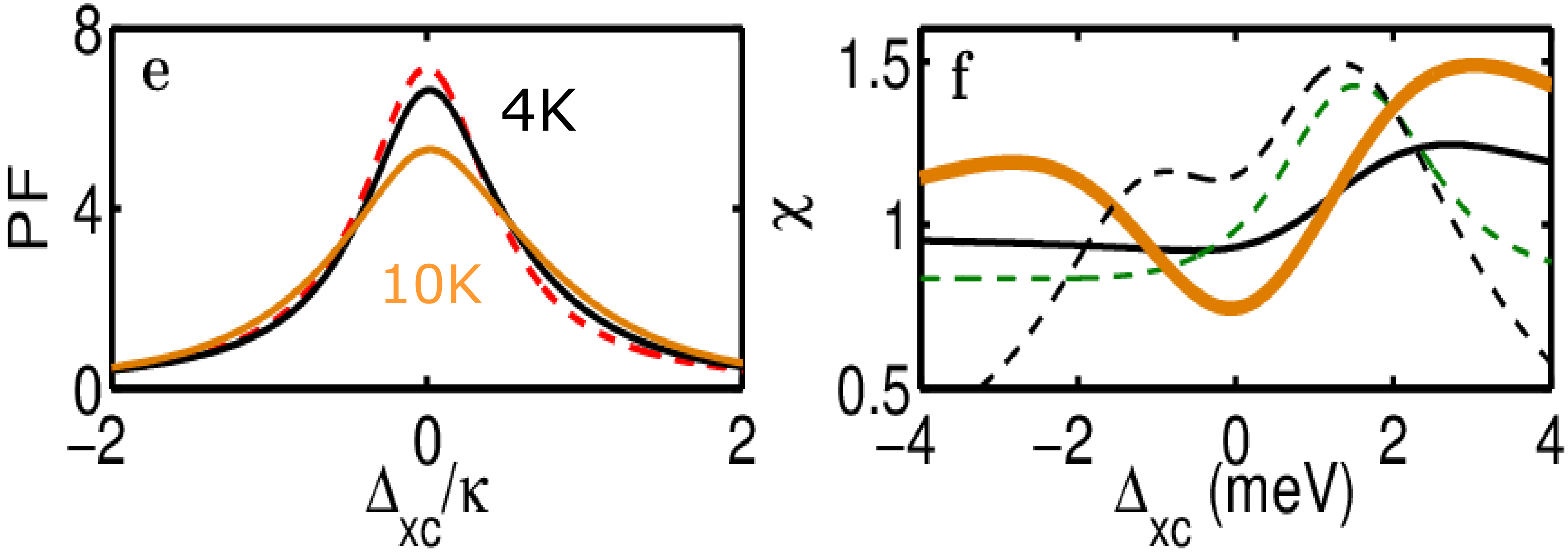
Phonon-Modified SE: Intermediate Q

$Q = 2300 \qquad \kappa = 0.6\, meV$

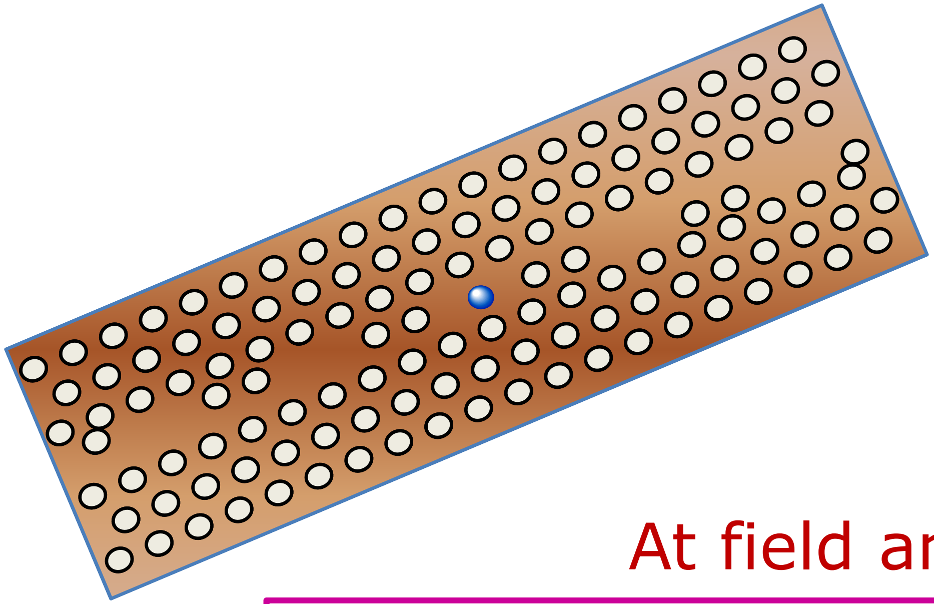


Cavity Results for Phonon-Modified SE

$Q = 600 \qquad \kappa = 2.4\,meV$



Example 2: CROW structure



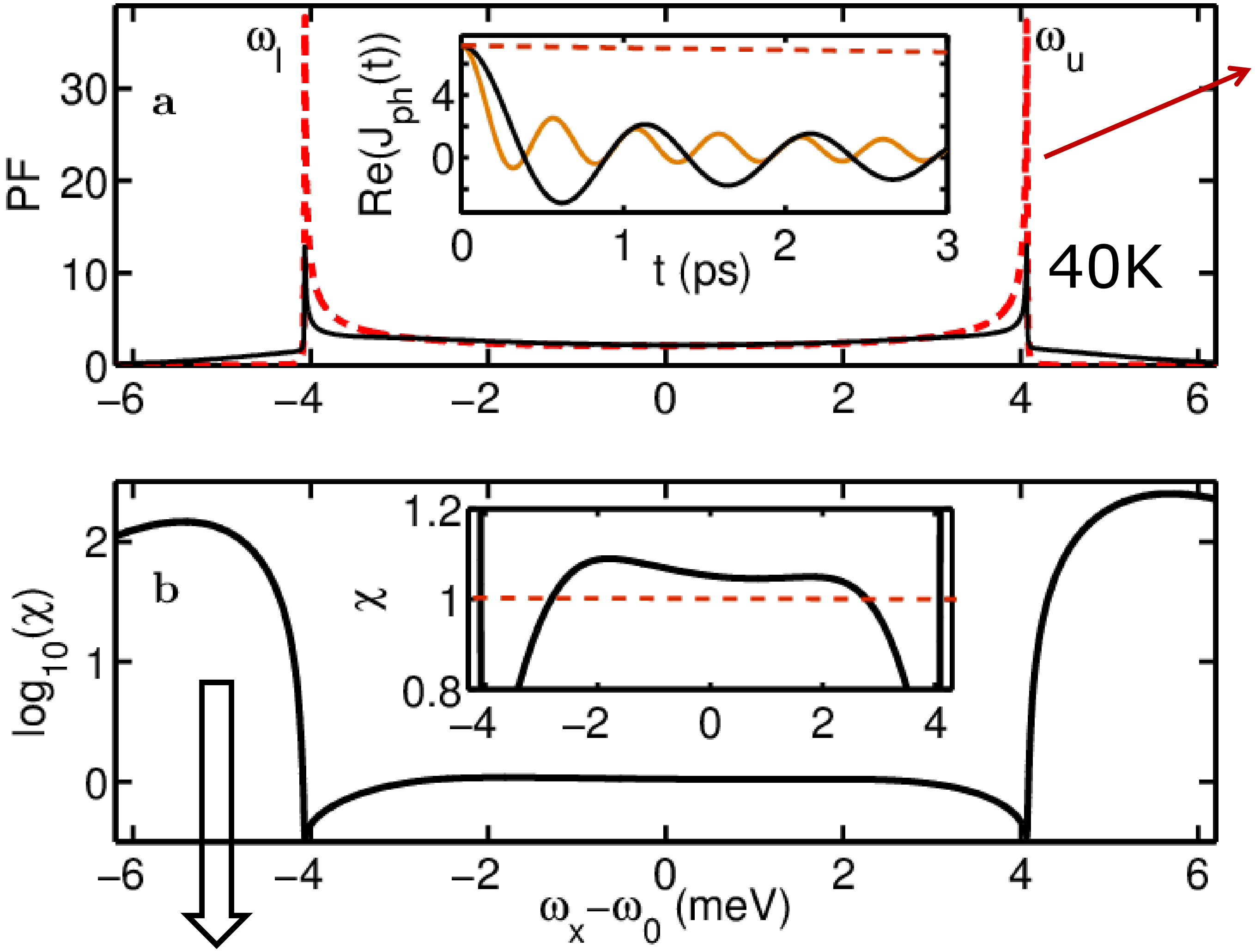
Regions of slow light
enhance the PF,
highly non-Lorentzian

At field antinode

$$\boxed{J_{\mathrm{ph}}(\omega) = \frac{-d^2\omega}{2\hbar\epsilon_0 n_b^2 V_{\mathrm{eff}}} \frac{1}{\pi} \mathrm{Im}\left[\frac{1}{\sqrt{(\omega-\tilde{\omega}_u)(\omega-\tilde{\omega}_l^*)}}\right]}$$

LDOS model details: Fussell, Hughes, Dignam, PRB (2007)

Phonon-modified SE from a CROW



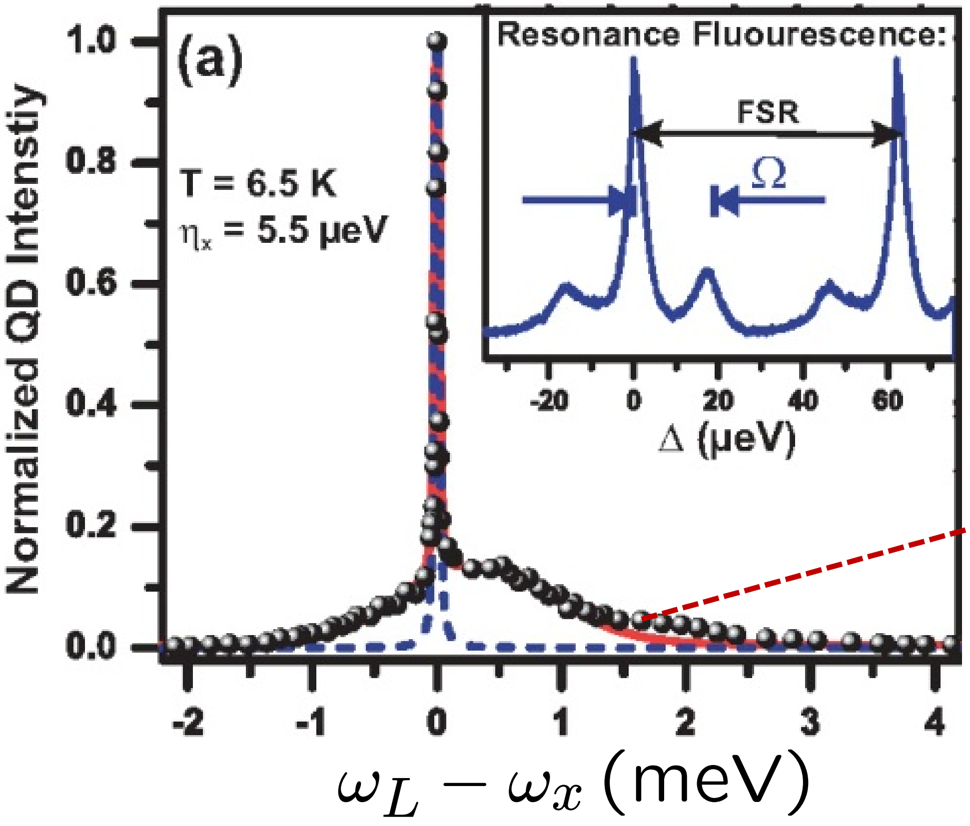
no
phonons

Two orders of magnitude SE increase outside band

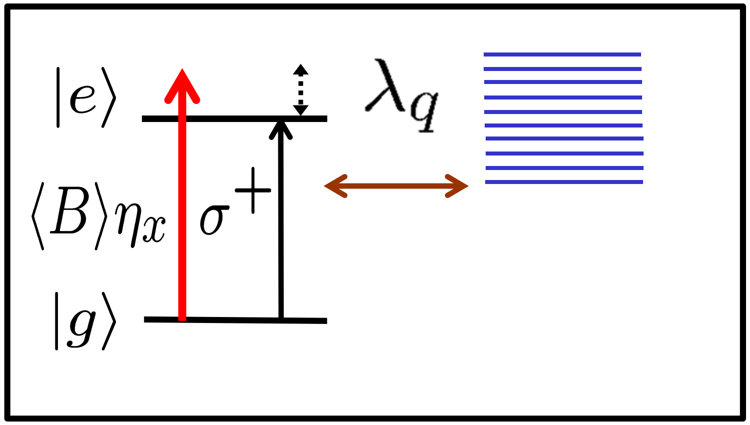
Coherent excitation: simple no-cavity case

$$\Gamma_{\rm ph}^{\sigma^+}(\Delta_{Lx},\eta_x) = 2\langle B\rangle^2 \eta_x^2 {\rm Re}\int_0^\infty d\tau e^{i\Delta_{Lx}\tau}\left[e^{\phi(\tau)}-1\right]$$

planar QD (InAs) sample



Roy and Hughes, PRX, 2012

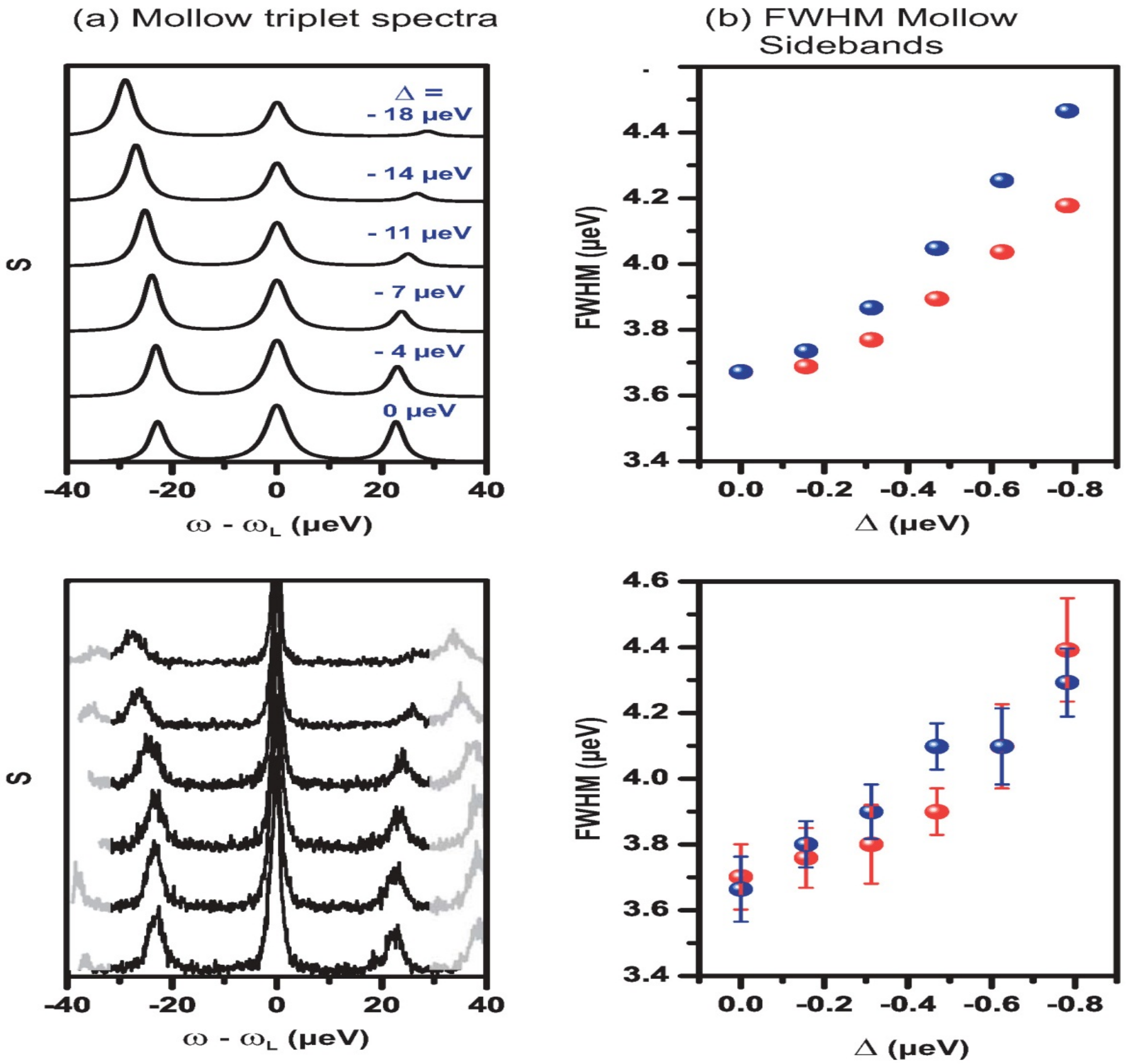


Polaron ME theory

$$\Gamma_{\rm ph}^{\sigma^+} \neq 0$$

S. Weiler et al, PRB 86, 241304(R) (2012).

Can also explain the Mollow Triplet Data


(a) Mollow triplet spectra

(b) FWHM Mollow Sidebands

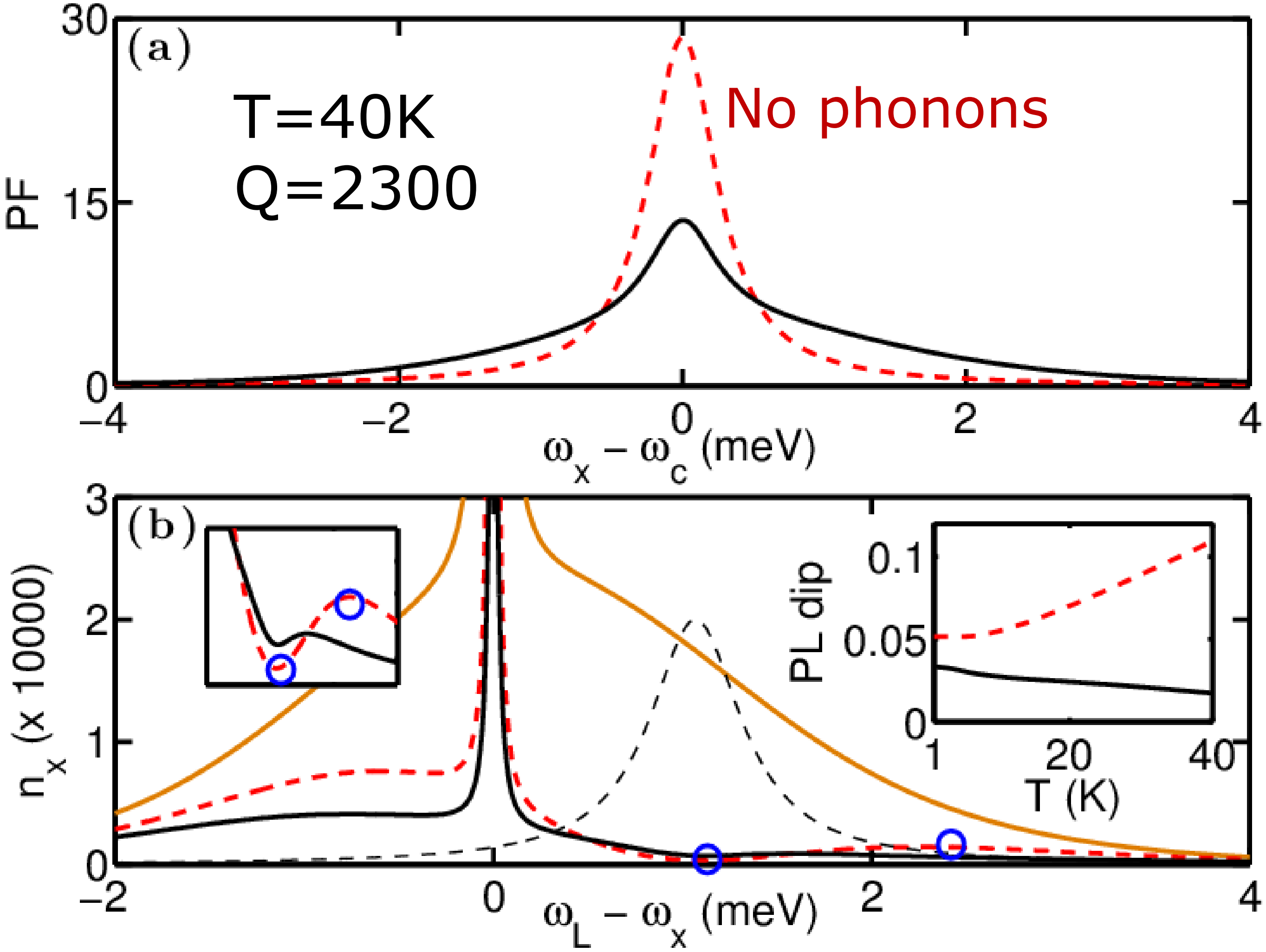
Ultaq et al., 2013
Opt. Express

Theory

T = 6 K

Experiment

QD coherently excited, Q=2300 cavity

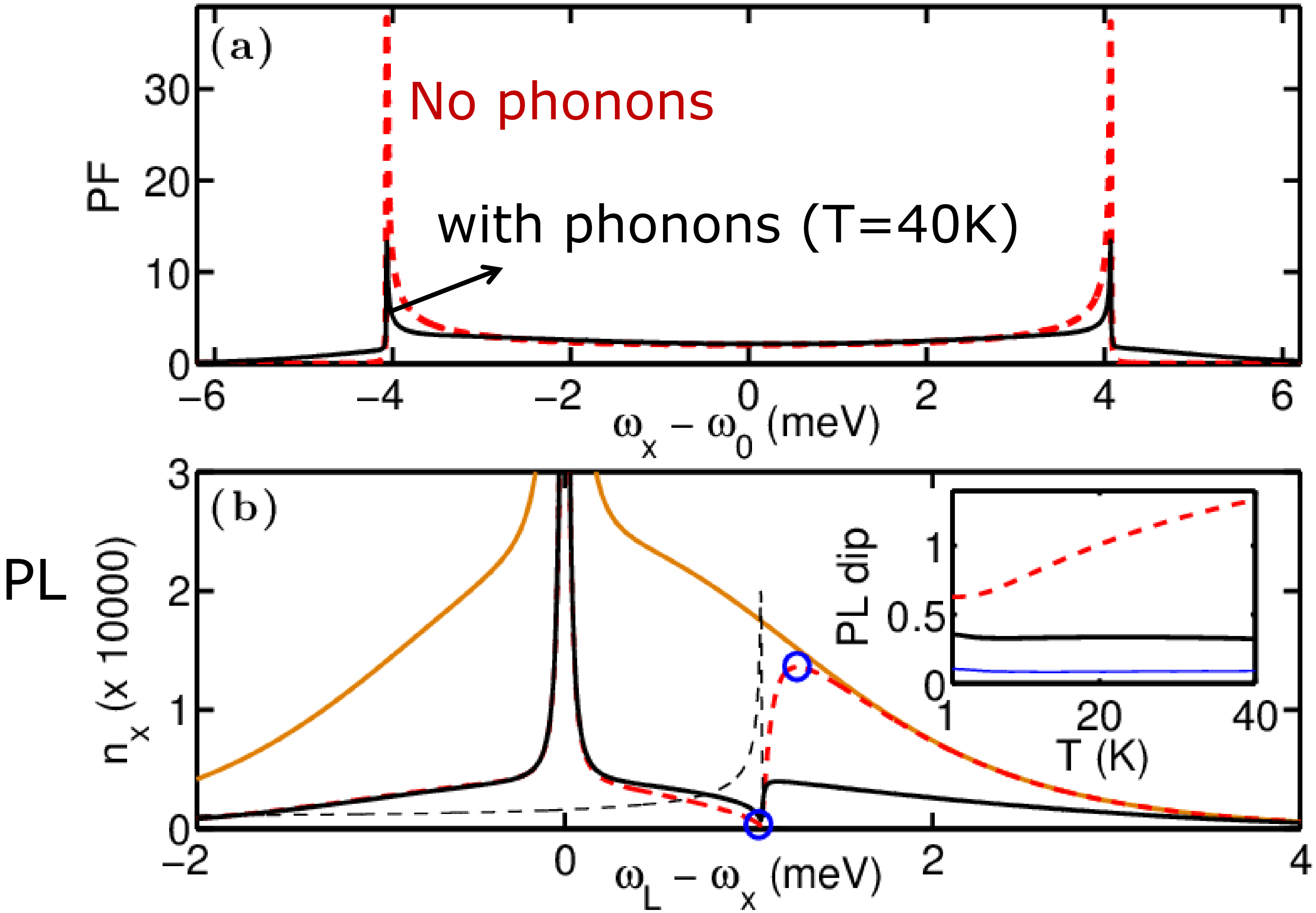


T=40K
Q=2300

No phonons

PL

QD coherently excited, CROW structure



Summary

- In presence of phonons, quantum dot SE rate in a structured photonic medium causes Fermi's golden rule to break down
- The breakdown is particularly significant in low to intermediate Q cavities and non-Lorentzian bath functions like waveguides
- Theory can be applied to any photon reservoir spectral function and effects should show up strongly in both PL and incoherent spectra

Refs: K. Roy-Choudhury and S. Hughes, arXiv:1406.3649 ; ibid, arXiv:1411.6050