# Real-Time Path Integrals for Laser Driven Carrier-Phonon Dynamics in Quantum Dots

Martin Glässl, Vollrath Martin Axt

Universität Bayreuth

08.11.2012

# Outline

Introduction

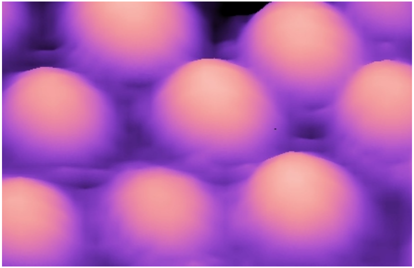
Path Integral Approach

Selected Results

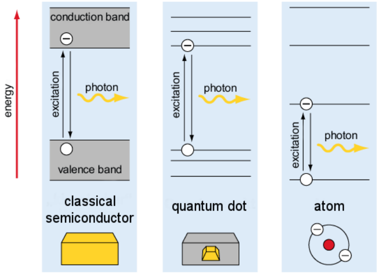
# Introduction

# Quantum Dots

## AFM image of quantum dots

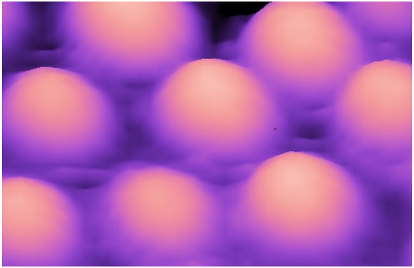


## Energy schemes in comparison

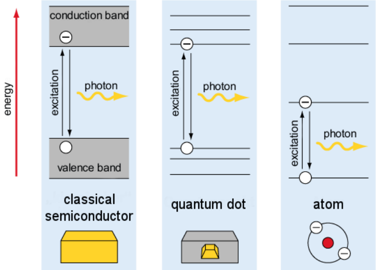


# Quantum Dots

AFM image of quantum dots



Energy schemes in comparison

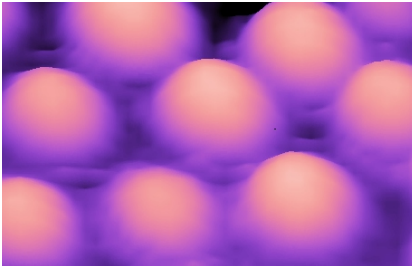



**Quantum dots are of high technological interest for various applications:**

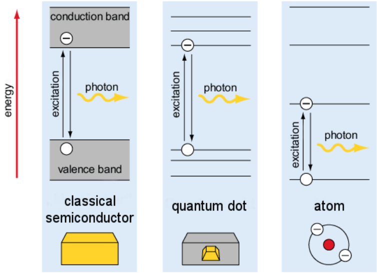
- single or entangled photon sources,
- new lasers,
- quantum information processing devices, ...

# Quantum Dots

AFM image of quantum dots



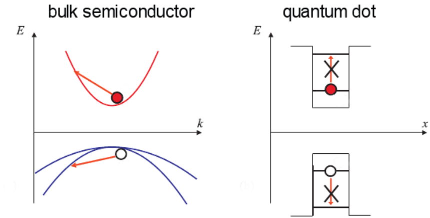
Energy schemes in comparison




**Quantum dots represent prototypes of quantum dissipative systems:**

$\Rightarrow$ Fascinating opportunities to study system-environment interactions

# Interaction with Phonons




**Discrete energy levels** $\Rightarrow$

- suppression of real transitions: "phonon bottleneck"
- elastic scattering processes: "pure dephasing"

## Pure dephasing coupling

- virtual transitions without change of occupations
- dominant dephasing mechanism in strongly confined quantum dots
- prototype of a non-Markovian interaction

# Model

## Hamiltonian for a laser-driven phonon-coupled L-level QD-system

$$H = \sum_{\nu} \hbar\omega_\nu |\nu\rangle\langle\nu| - \sum_{\nu\nu'} \hbar M_{\nu\nu'} |\nu\rangle\langle\nu'| + \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}}\, b_{\mathbf{q}}^\dagger b_{\mathbf{q}} + \sum_{\mathbf{q}\nu} \hbar\left(\gamma_{\mathbf{q}}^{\nu} b_{\mathbf{q}} + \gamma_{\mathbf{q}}^{\nu *} b_{\mathbf{q}}^\dagger\right) |\nu\rangle\langle\nu|$$

- Electronic system comprises L levels
- **M**: Matrix of dipole interactions with a classical light field
- $b_{\mathbf{q}}^\dagger$: creation operator of a phonon with wave vector **q** and energy $\hbar\omega_{\mathbf{q}}$
- $\gamma_{\mathbf{q}}^{\nu}$: carrier-phonon coupling constants (pure dephasing processes)
- GaAs: deformation potential coupling to LA phonons dominates

# Model

## Hamiltonian for a laser-driven phonon-coupled L-level QD-system

$$H = \sum_{\nu} \hbar\omega_\nu |\nu\rangle\langle\nu| - \sum_{\nu\nu'} \hbar M_{\nu\nu'} |\nu\rangle\langle\nu'| + \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}}\, b^\dagger_{\mathbf{q}} b_{\mathbf{q}} + \sum_{\mathbf{q}\nu} \hbar\big(\gamma^\nu_{\mathbf{q}} b_{\mathbf{q}} + \gamma^{\nu *}_{\mathbf{q}} b^\dagger_{\mathbf{q}}\big)|\nu\rangle\langle\nu|$$

**Aim:** Calculate dynamics of the electronic subsystem

- Analytical solutions are only known for limiting cases
- Usually: Treatment of the carrier-phonon coupling within approximations
  $\Rightarrow$ Validity of results is unclear
- Here: Real-time path integrals: no approximations!

# Path Integral Approach

# Path Integral Representation

- Reduced electronic density matrix:

$$\widehat{\overline{\rho}}(t) = \mathrm{Tr}_{ph}\left[\widehat{U}(t)\widehat{\rho}(0)\widehat{U}^{\dagger}(t)\right], \qquad \text{where } \widehat{U}(t) = \widehat{T}\exp\left(\tfrac{i}{\hbar}\int_0^t \widehat{H}(\tau)d\tau\right)$$

# Path Integral Representation

- Reduced electronic density matrix:
  $\widehat{\overline{\rho}}(t) = \mathrm{Tr}_{ph}\left[\widehat{U}(t)\widehat{\rho}(0)\widehat{U}^{\dagger}(t)\right]$, where $\widehat{U}(t) = \widehat{T}\exp\left(\frac{i}{\hbar}\int_0^t \widehat{H}(\tau)d\tau\right)$
- Discretize time evolution operator ($t_n = n\varepsilon$):
  $\widehat{U}(t_N) \approx e^{-i\epsilon\widehat{H}(t_N)/\hbar}\ e^{-i\epsilon\widehat{H}(t_{N-1})/\hbar}\ \ldots\ e^{-i\epsilon\widehat{H}(t_1)/\hbar}$

# Path Integral Representation

- Reduced electronic density matrix:
  $\widehat{\overline{\rho}}(t) = \mathrm{Tr}_{ph}\left[\widehat{U}(t)\widehat{\rho}(0)\widehat{U}^{\dagger}(t)\right], \qquad \text{where } \widehat{U}(t) = \widehat{T}\exp\left(\frac{i}{\hbar}\int_0^t \widehat{H}(\tau)d\tau\right)$
- Discretize time evolution operator ($t_n = n\varepsilon$):
  $\widehat{U}(t_N) \approx e^{-i\epsilon\widehat{H}(t_N)/\hbar}\; e^{-i\epsilon\widehat{H}(t_{N-1})/\hbar}\; \ldots\; e^{-i\epsilon\widehat{H}(t_1)/\hbar}$
- Insert identity-operators $\widehat{I}_n = \left(\sum_{\alpha_n} |\alpha_n\rangle\langle\alpha_n|\right) \otimes \left(\int d\mu_n\, |\mathcal{Z}_n\rangle\langle\mathcal{Z}_n|\right)$:
  $\widehat{U}(t_N) \approx e^{-i\epsilon\widehat{H}(t_N)/\hbar}\, \widehat{I}_{N-1}\; \ldots\; \widehat{I}_1\; e^{-i\epsilon\widehat{H}(t_1)/\hbar}$

# Path Integral Representation

- Reduced electronic density matrix:
  $\widehat{\overline{\rho}}(t) = \mathrm{Tr}_{ph}\left[\widehat{U}(t)\widehat{\rho}(0)\widehat{U}^{\dagger}(t)\right]$, where $\widehat{U}(t) = \widehat{T}\exp\left(\frac{i}{\hbar}\int_0^t \widehat{H}(\tau)d\tau\right)$
- Discretize time evolution operator ($t_n = n\varepsilon$):
  $\widehat{U}(t_N) \approx e^{-i\epsilon\widehat{H}(t_N)/\hbar}\; e^{-i\epsilon\widehat{H}(t_{N-1})/\hbar}\; \ldots\; e^{-i\epsilon\widehat{H}(t_1)/\hbar}$
- Insert identity-operators $\widehat{I}_n = \left(\sum_{\alpha_n}|\alpha_n\rangle\langle\alpha_n|\right)\otimes\left(\int d\mu_n\,|\mathcal{Z}_n\rangle\langle\mathcal{Z}_n|\right)$:
  $\widehat{U}(t_N) \approx e^{-i\epsilon\widehat{H}(t_N)/\hbar}\,\widehat{I}_{N-1}\;\ldots\;\widehat{I}_1\; e^{-i\epsilon\widehat{H}(t_1)/\hbar}$
- Integrate out phonon degrees of freedom

# Path Integral Representation

- Reduced electronic density matrix:
  $\widehat{\overline{\rho}}(t) = \mathrm{Tr}_{ph}\left[\widehat{U}(t)\widehat{\rho}(0)\widehat{U}^{\dagger}(t)\right]$, where $\widehat{U}(t) = \widehat{T}\exp\left(\frac{i}{\hbar}\int_0^t \widehat{H}(\tau)d\tau\right)$
- Discretize time evolution operator ($t_n = n\varepsilon$):
  $\widehat{U}(t_N) \approx e^{-i\epsilon\widehat{H}(t_N)/\hbar}\; e^{-i\epsilon\widehat{H}(t_{N-1})/\hbar}\; \dots\; e^{-i\epsilon\widehat{H}(t_1)/\hbar}$
- Insert identity-operators $\widehat{I}_n = \left(\sum_{\alpha_n} |\alpha_n\rangle\langle\alpha_n|\right) \otimes \left(\int d\mu_n\, |\mathcal{Z}_n\rangle\langle\mathcal{Z}_n|\right)$:
  $\widehat{U}(t_N) \approx e^{-i\epsilon\widehat{H}(t_N)/\hbar}\, \widehat{I}_{N-1}\; \dots\; \widehat{I}_1\; e^{-i\epsilon\widehat{H}(t_1)/\hbar}$
- Integrate out phonon degrees of freedom

$$\Rightarrow \overline{\rho}_{\alpha_N\beta_N} = e^{i\Omega t(\beta_N-\alpha_N)} \sum_{\{\alpha_n,\beta_n\}} \prod_{n=1}^{N} M_{\alpha_n}^{\alpha_{n-1}} M_{\beta_n}^{\beta_{n-1}*} \times \prod_{n'=1}^{n} e^{\mathcal{S}_{nn'}}\; \overline{\rho}_{\alpha_0\beta_0}(0)$$

Influence functional $\mathcal{S}_{nn'} = -\left[\zeta(\alpha_n) - \zeta(\beta_n)\right]\left[K_{n-n'}\zeta(\alpha_{n'}) - K^*_{n-n'}\zeta(\beta_{n'})\right]$

# Memory Kernel & Memory Truncation Scheme

$$K_{n\neq 0} = \int_{(n-1)\epsilon}^{n\epsilon} d\tau \int_0^{\epsilon} d\tau' \Gamma(\tau - \tau') \qquad \text{and} \qquad K_0 = \int_0^{\epsilon} d\tau \int_0^{\tau} d\tau' \Gamma(\tau - \tau')$$

$$\Gamma(t) = \int_0^{\infty} d\omega J(\omega) \left[ \cos(\omega t) \coth\left(\frac{\hbar\omega}{2k_B T}\right) - i \sin(\omega t) \right]$$

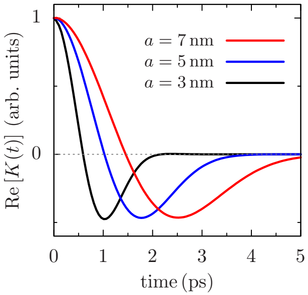
Spectral density $J(\omega)$ is of the superohmic coupling type: $J(\omega) \propto \omega^3$ for $\omega \to 0$

# Memory Kernel & Memory Truncation Scheme

$$K_{n\neq 0} = \int_{(n-1)\epsilon}^{n\epsilon} d\tau \int_0^{\epsilon} d\tau' \Gamma(\tau - \tau') \quad \text{and} \quad K_0 = \int_0^{\epsilon} d\tau \int_0^{\tau} d\tau' \Gamma(\tau - \tau')$$

$$\Gamma(t) = \int_0^{\infty} d\omega J(\omega) \left[\cos(\omega t) \coth\left(\frac{\hbar\omega}{2k_B T}\right) - i \sin(\omega t)\right]$$

Spectral density $J(\omega)$ is of the superohmic coupling type: $J(\omega) \propto \omega^3$ for $\omega \rightarrow 0$




- $K_n$ tends sufficiently fast to zero
- $\Rightarrow$ Memory truncation:

  Choose a cutoff $n_c$ such that $K_{n>n_c} = 0$
- Enables calculations for arbitrarily long times

# Augmented Density Matrix & Paths

$$\Rightarrow \overline{\rho}_{\alpha_N\beta_N} = e^{i\Omega t(\beta_N-\alpha_N)} \sum_{\{\alpha_n,\beta_n\}} \prod_{n=1}^{N} M_{\alpha_n}^{\alpha_{n-1}} M_{\beta_n}^{\beta_{n-1}*} \times \prod_{n'=1}^{n} \quad e^{\mathcal{S}_{nn'}} \; \overline{\rho}_{\alpha_0\beta_0}(0)$$

# Augmented Density Matrix & Paths

$$\Rightarrow \overline{\rho}_{\alpha_N\beta_N} = e^{i\Omega t(\beta_N-\alpha_N)} \sum_{\{\alpha_n,\beta_n\}} \prod_{n=1}^{N} M_{\alpha_n}^{\alpha_{n-1}} M_{\beta_n}^{\beta_{n-1}*} \times \prod_{n'=n-n_c}^{n} e^{\mathcal{S}_{nn'}} \; \overline{\rho}_{\alpha_0\beta_0}(0)$$

# Augmented Density Matrix & Paths

$$\Rightarrow \overline{\rho}_{\alpha_N\beta_N} = e^{i\Omega t(\beta_N-\alpha_N)} \sum_{\{\alpha_n,\beta_n\}} \prod_{n=1}^{N} M_{\alpha_n}^{\alpha_{n-1}} M_{\beta_n}^{\beta_{n-1}*} \times \prod_{n'=n-n_c}^{n} e^{\mathcal{S}_{nn'}} \; \overline{\rho}_{\alpha_0\beta_0}(0)$$

- Introduce **augmented density matrix R**:

  (N. Makri and D. Makarov, J. Chem. Phys. **102**, 4600 (1995))

  $R_n = T_{n,\dots,n-n_c-1}\, R_{n-1}$ Recurrence without memory

- Augmented density matrix is given as a function of **paths**, where at time step n each path is given as a sequence of the form $(\alpha_n, \dots, \alpha_{n-n_c}, \beta_n, \dots, \beta_{n-n_c})$

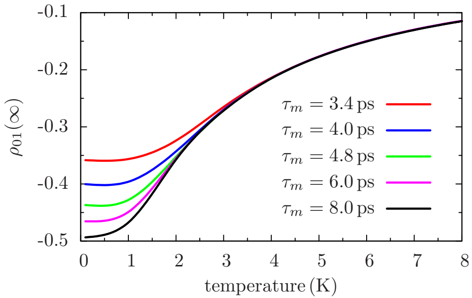
⇒ Each path is a sequence of the length $2(n_c + 1)$

⇒ For a L-level system, there are $L^{2(n_c+1)}$ possible paths

# Memory Depth & Convergence Properties

Memory depth is given by $n_c\,\varepsilon$ and has to be chosen sufficiently long:

**Example:** Temperature-dependence of the stationary offdiagonal element $\rho_{01}$




Long memory depths for

- low temperatures and
- weak fields

M. Glässl et al., PRB **84**, 195311 (2011)

# Specifics due to the superohmic Coupling

Comparison with exact long time asymptotics yields the constraint:

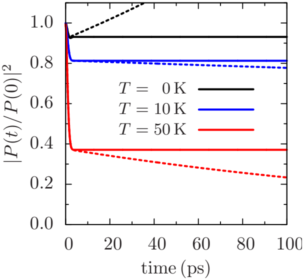
$$\sum_{n=0}^{n_c} \mathrm{Re}[K_n] = 0$$

# Specifics due to the superohmic Coupling

Comparison with exact long time asymptotics yields the constraint:

$$\sum_{n=0}^{n_c} \mathrm{Re}[K_n] = 0$$

**Example:** Time dependence of the optical polarization after an ultrafast pulse




- Numerical results with enforcing the constraint are indistinguishable from analytical results
- Simulations without this constraint show qualitatively different long time asymptotics

A. Vagov et al., PRB **83**, 094303 (2011)

# Correlation Expansion

**Quantum kinetic density matrix approach:**

- Set up Heisenberg equations of motion for $\langle dc \rangle$ and $\langle c^\dagger c \rangle$
- Equations contain single phonon assisted density matrices like $\langle c^\dagger c\, b_{\mathbf{q}} \rangle$
- Equations for single assisted quantities contain double assisted quantities, ...

  $\Rightarrow$ Infinite hierarchy of higher-order density matrix elements

  $\Rightarrow$ Truncate hierarchy by factorizing higher order terms on a chosen level

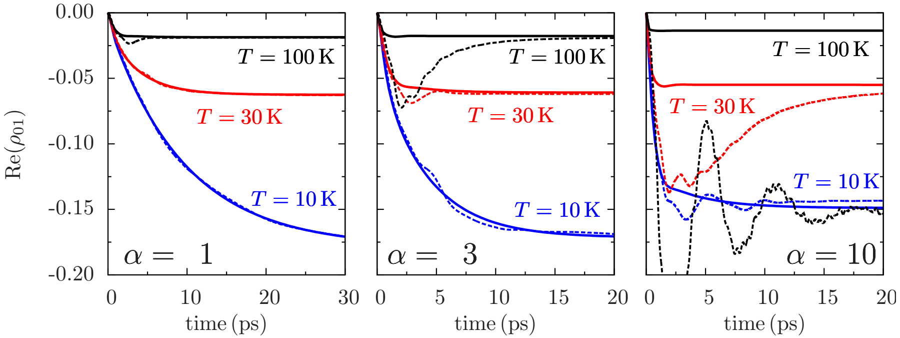
**4th-Order correlation expansion:**

- Include all quantities with up to four operators like $\langle c^\dagger c\, b_{\mathbf{q}} b_{\mathbf{q}'} \rangle$
- Factorize higher order assisted density matrices like $\langle c^\dagger c\, b_{\mathbf{q}} b_{\mathbf{q}'} b_{\mathbf{q}''} \rangle$

F. Rossi and T. Kuhn, Rev. Mod. Phys. **74**, 895 (2002)

# Path Integrals vs. 4th-Order Correlation Expansion

Increase coupling constants $|\gamma_q|^2$ by a factor $\alpha$.

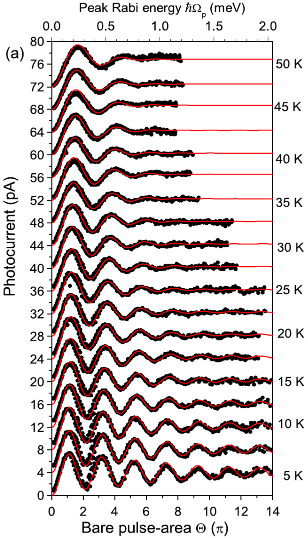



**Correlation expansion breaks down at strong couplings and/or high T**

M. Glässl et al., PRB **84**, 195311 (2011)

# Selected Results

# Experimental Results: Pulsed Excitation




A. J. Ramsay et al., PRL **104**, 017402 (2010)

A. J. Ramsay et al., PRL **105**, 177402 (2010)

Damped oscillations with renormalized period

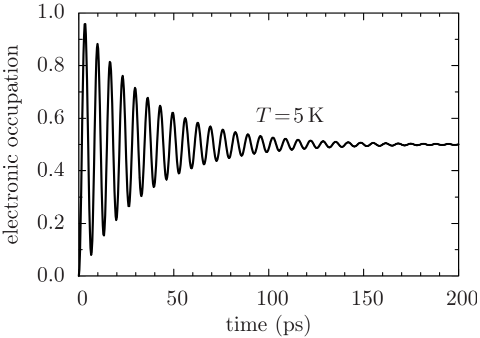
**Excellent agreement with theoretical predictions!**

J. Förstner et al., PRL **91**, 127401 (2003)

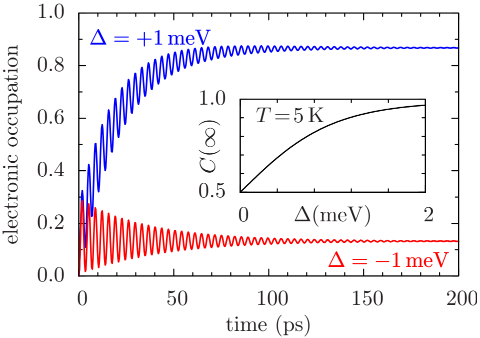
A. Krügel et al., Appl. Phys. B: Lasers Opt. **81**, 897 (2005)

A. Vagov et al., PRL **98**, 227403 (2007)

# Constant Driving: Stationary Nonequilibrium



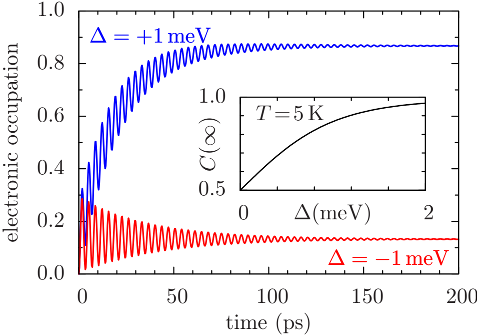
# Constant Driving: Stationary Nonequilibrium




**Long-time dynamics:**

- positive detunings:
  stationary occupation $> 0.5$
- negative detunings:
  stationary occupation $< 0.5$

# Constant Driving: Stationary Nonequilibrium




**Long-time dynamics:**

- positive detunings: stationary occupation $> 0.5$
- negative detunings: stationary occupation $< 0.5$

**What characterizes the stationary nonequilibrium state?**

M. Glässl et al., PRB **84**, 195311 (2011)

# Quantized Light Fields: Revival Dynamics

Coupling to a quantized light field instead of coupling to a classical field:

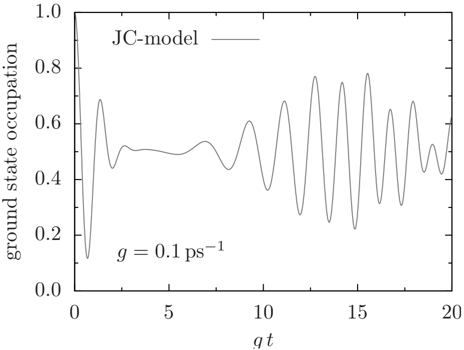
$$H = \hbar\omega_x |X\rangle\langle X| - \hbar g \left( a^\dagger |0\rangle\langle X| + a|X\rangle\langle 0| \right) + \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} b_{\mathbf{q}}^\dagger b_{\mathbf{q}} + \sum_{\mathbf{q}} \hbar \left( \gamma_{\mathbf{q}} b_{\mathbf{q}} + \gamma_{\mathbf{q}} b_{\mathbf{q}}^\dagger \right) |X\rangle\langle X|$$

Coupling to a quantized light field instead of coupling to a classical field:

$$H = \hbar\omega_x |X\rangle\langle X| - \hbar g \left( a^\dagger |0\rangle\langle X| + a|X\rangle\langle 0| \right) + \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} b^\dagger_{\mathbf{q}} b_{\mathbf{q}} + \sum_{\mathbf{q}} \hbar \left( \gamma_{\mathbf{q}} b_{\mathbf{q}} + \gamma_{\mathbf{q}} b^\dagger_{\mathbf{q}} \right) |X\rangle\langle X|$$

Initial cavity preparation:

coherent state with $\langle n \rangle = 5$



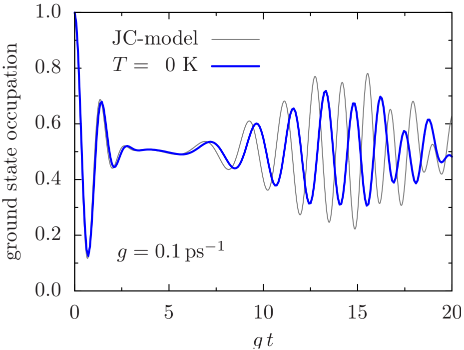
# Quantized Light Fields: Revival Dynamics

Coupling to a quantized light field instead of coupling to a classical field:

$$H = \hbar\omega_x |X\rangle\langle X| - \hbar g \left( a^\dagger |0\rangle\langle X| + a|X\rangle\langle 0| \right) + \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} b^\dagger_{\mathbf{q}} b_{\mathbf{q}} + \sum_{\mathbf{q}} \hbar \left( \gamma_{\mathbf{q}} b_{\mathbf{q}} + \gamma_{\mathbf{q}} b^\dagger_{\mathbf{q}} \right) |X\rangle\langle X|$$

Initial cavity preparation:

coherent state with $\langle n \rangle = 5$



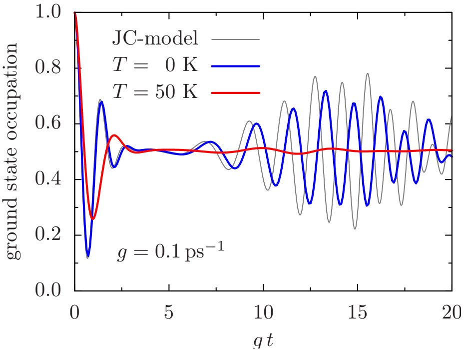
# Quantized Light Fields: Revival Dynamics

Coupling to a quantized light field instead of coupling to a classical field:

$$H = \hbar\omega_x |X\rangle\langle X| - \hbar g\left(a^\dagger|0\rangle\langle X| + a|X\rangle\langle 0|\right) + \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} b_{\mathbf{q}}^\dagger b_{\mathbf{q}} + \sum_{\mathbf{q}} \hbar\left(\gamma_{\mathbf{q}} b_{\mathbf{q}} + \gamma_{\mathbf{q}} b_{\mathbf{q}}^\dagger\right)|X\rangle\langle X|$$

Initial cavity preparation:

coherent state with $\langle n \rangle = 5$



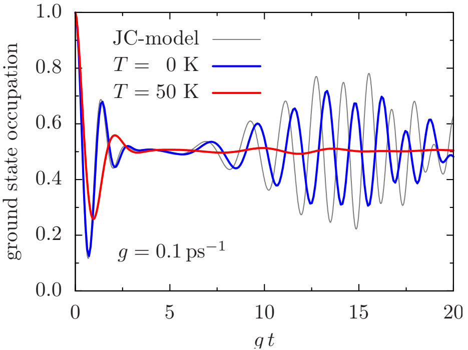
# Quantized Light Fields: Revival Dynamics

Coupling to a quantized light field instead of coupling to a classical field:

$$H = \hbar\omega_x |X\rangle\langle X| - \hbar g \left( a^\dagger |0\rangle\langle X| + a|X\rangle\langle 0| \right) + \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} b^\dagger_{\mathbf{q}} b_{\mathbf{q}} + \sum_{\mathbf{q}} \hbar \left( \gamma_{\mathbf{q}} b_{\mathbf{q}} + \gamma_{\mathbf{q}} b^\dagger_{\mathbf{q}} \right) |X\rangle\langle X|$$

Initial cavity preparation:

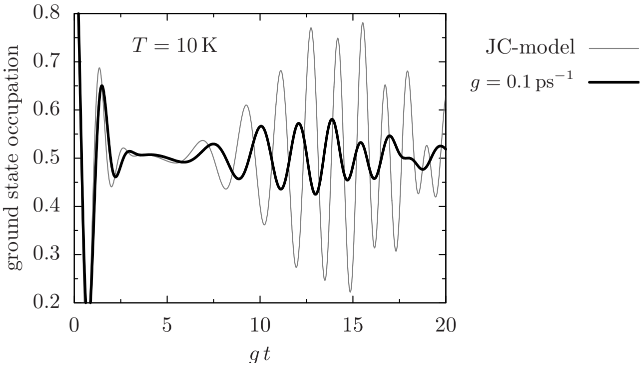
coherent state with $\langle n \rangle = 5$




**Acoustic phonon coupling strongly affects the dynamics, even at $\mathbf{T} = 0$.**

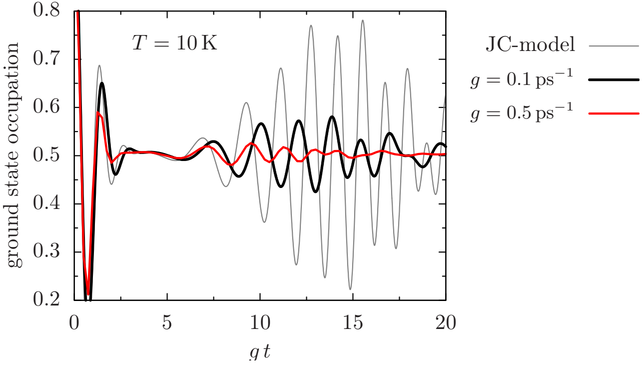
# Revivals for Different Light-Matter Coupling Strengths

Initial cavity preparation: coherent state with $\langle n \rangle = 5$



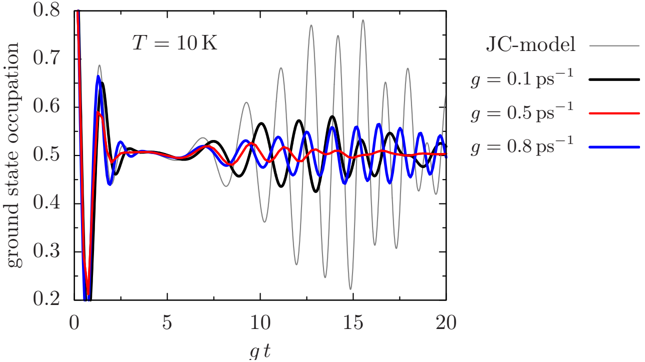
# Revivals for Different Light-Matter Coupling Strengths

Initial cavity preparation: coherent state with $\langle n \rangle = 5$



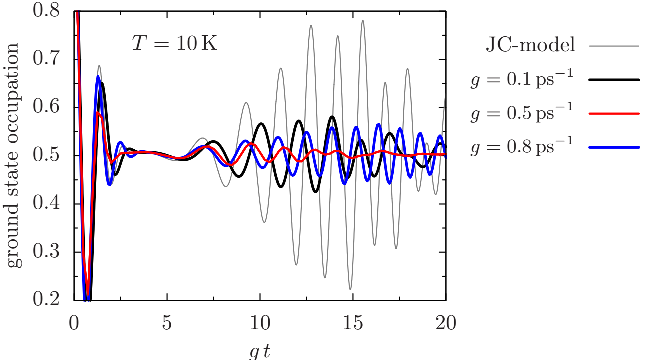
# Revivals for Different Light-Matter Coupling Strengths

Initial cavity preparation: coherent state with $\langle n \rangle = 5$



# Revivals for Different Light-Matter Coupling Strengths

Initial cavity preparation: coherent state with $\langle n \rangle = 5$

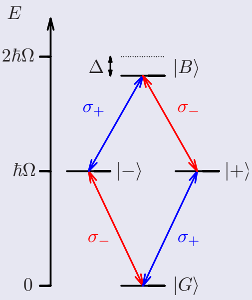



**A stronger light-matter coupling reduces the visibility of the revival for parameters usually accessible in experiments.**

M. Glässl et al., PRB **86**, 035319 (2012)

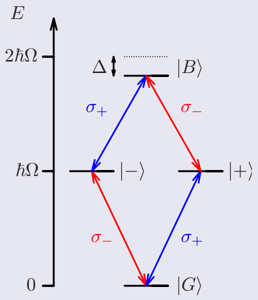
# Exciton-Biexciton System: Relaxation Dynamics

## Energy level sketch



# Exciton-Biexciton System: Relaxation Dynamics

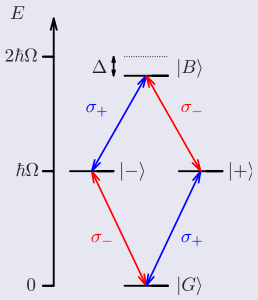
## Energy level sketch



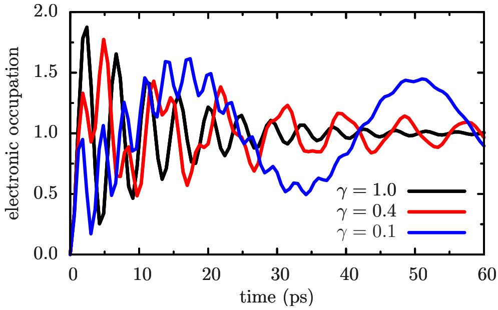

Polarization parameter $\gamma = f_{\sigma_+}/f_{\sigma_-}$

# Exciton-Biexciton System: Relaxation Dynamics

## Energy level sketch




Polarization parameter $\gamma = f_{\sigma_+}/f_{\sigma_-}$




**Phonon induced damping strongly depends on the polarization although the carrier-phonon interaction is spin-independent!**

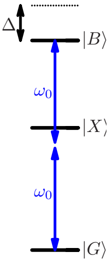
M. Glässl et al., PRB **85**, 195306 (2012), M. Glässl et al., to be published

# Robust Biexciton Preparation via Adiabatic Rapid Passage

Excitation with a linearly polarized frequency-swept Gaussian pulse (chirp $\alpha$):

$$dE = \frac{A}{\sqrt{2\pi\tau\tau_0}} \exp(-\frac{t^2}{2\tau^2}) \exp(-it(\omega_0 + at)) \quad \text{with} \quad a = \alpha/(\alpha^2 + \tau_0^4)$$

$$\tau = \sqrt{\alpha^2/\tau_0^2 + \tau_0^2}$$

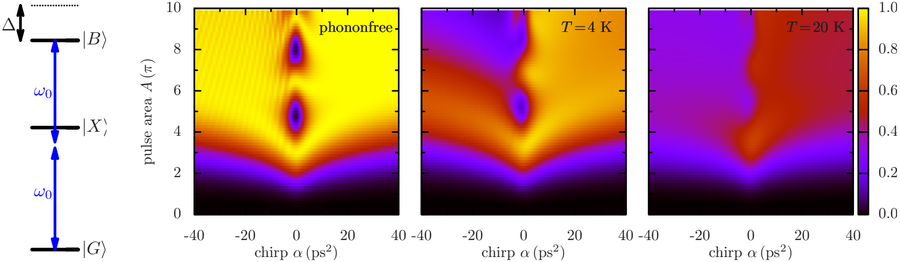


# Robust Biexciton Preparation via Adiabatic Rapid Passage

Excitation with a linearly polarized frequency-swept Gaussian pulse (chirp $\alpha$):

$$dE = \frac{A}{\sqrt{2\pi\tau\tau_0}} \exp(-\frac{t^2}{2\tau^2}) \exp(-it(\omega_0 + at)) \quad \text{with} \quad a = \alpha/(\alpha^2 + \tau_0^4)$$

$$\tau = \sqrt{\alpha^2/\tau_0^2 + \tau_0^2}$$

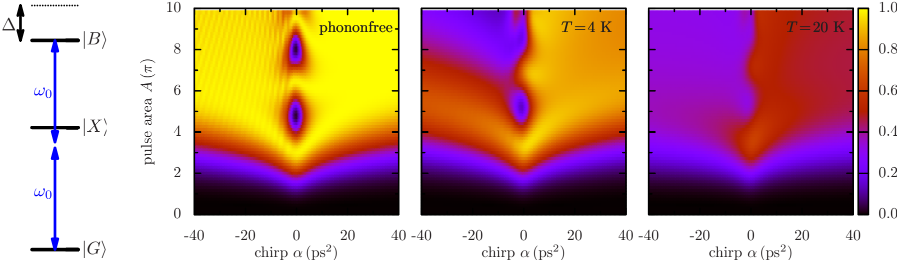


# Robust Biexciton Preparation via Adiabatic Rapid Passage

Excitation with a linearly polarized frequency-swept Gaussian pulse (chirp $\alpha$):

$$dE = \frac{A}{\sqrt{2\pi\tau\tau_0}} \exp(-\frac{t^2}{2\tau^2}) \exp(-it(\omega_0 + at)) \quad \text{with} \quad a = \alpha/(\alpha^2 + \tau_0^4)$$

$$\tau = \sqrt{\alpha^2/\tau_0^2 + \tau_0^2}$$




**High fidelity preparation only for low temperatures and positive chirps.**

M. Glässl et al., to be published

# Summary

- Acoustic phonons strongly affect the dynamics of driven quantum dots
- Phonons mostly hinder but can sometimes also enable control schemes

- Real-time path integrals allow for numerically exact calculations
- Method is applicable for an almost unlimited parameter range
- Low-temperature studies are numerically most demanding
- Numerical effort rises drastically with the number of electronic levels
- Superohmic coupling requires a special treatment