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

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Introduction
Quantum Dots

AFM image of quantum dots

Energy schemes in comparison

Martin Glässl (Universität Bayreuth)
Quantum dots are of high technological interest for various applications:

- single or entangled photon sources,
- new lasers,
- quantum information processing devices, ...
Quantum dots represent prototypes of quantum dissipative systems:

⇒ 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

\[ H = \sum_\nu \hbar \omega_\nu |\nu\rangle\langle\nu| - \sum_{\nu\nu'} \hbar M_{\nu\nu'} |\nu\rangle\langle\nu'| + \sum_\mathbf{q} \hbar \omega_q b_\mathbf{q}^\dagger b_\mathbf{q} + \sum_\mathbf{q}\nu \hbar (\gamma^\nu_\mathbf{q} b_\mathbf{q} + \gamma^{\nu*}_\mathbf{q} b_\mathbf{q}^\dagger) |\nu\rangle\langle\nu| \]

- 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 \( \mathbf{q} \) and energy \( \hbar \omega_\mathbf{q} \)
- **\gamma^\nu_\mathbf{q}**: carrier-phonon coupling constants (pure dephasing processes)
- GaAs: deformation potential coupling to LA phonons dominates
Hamiltonian for a laser-driven phonon-coupled L-level QD-system

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**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:

\[ \hat{\rho}(t) = \text{Tr}_{ph} \left[ \hat{U}(t)\hat{\rho}(0)\hat{U}^\dagger(t) \right], \quad \text{where} \quad \hat{U}(t) = \hat{T} \exp \left( \frac{i}{\hbar} \int_0^t \hat{H}(\tau) d\tau \right) \]
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- Discretize time evolution operator \((t_n = n\varepsilon)\):
  \[
  \hat{U}(t_N) \approx e^{-i\varepsilon \hat{H}(t_N)/\hbar} \ e^{-i\varepsilon \hat{H}(t_{N-1})/\hbar} \ldots \ e^{-i\varepsilon \hat{H}(t_1)/\hbar}
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- Insert identity-operators \( \hat{I}_n = (\sum_{\alpha_n} \ket{\alpha_n}\bra{\alpha_n}) \otimes (\int d\mu_n \ket{Z_n}\bra{Z_n}) \):
  \[ \hat{U}(t_N) \approx e^{-i\varepsilon \hat{H}(t_N)/\hbar} \hat{I}_{N-1} \ldots \hat{I}_1 e^{-i\varepsilon \hat{H}(1)/\hbar} \]
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- Integrate out phonon degrees of freedom
Path Integral Representation

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  \[ \hat{U}(t_N) \approx e^{-i\varepsilon \hat{H}(t_N)/\hbar} \hat{I}_{N-1} \ldots \hat{I}_1 e^{-i\varepsilon \hat{H}(t_1)/\hbar} \]

- Integrate out phonon degrees of freedom

\[ \Rightarrow \quad \hat{\rho}_{\alpha_N\beta_N} = e^{i\Omega t(\beta_N - \alpha_N)} \sum_\{\alpha_n,\beta_n\} \prod_{n=1}^N M_{\alpha_n}^{\alpha_{n-1}} M_{\beta_n}^{\beta_{n-1}} \times \prod_{n'=1}^n e^{S_{nn'}} \hat{\rho}_{\alpha_0\beta_0}(0) \]

Influence functional \(S_{nn'} = -[\zeta(\alpha_n) - \zeta(\beta_n)][K_{n-n'}\zeta(\alpha_{n'}) - K_{n-n'}^*\zeta(\beta_{n'})] \)
Memory Kernel & Memory Truncation Scheme

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

\[ \Gamma(t) = \int_{0}^{\infty} d\omega J(\omega) \left[ \cos(\omega t) \coth \left( \frac{\hbar \omega}{2 k_B T} \right) - i \sin(\omega t) \right] \]

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

\[ \Rightarrow \bar{\rho}_{\alpha_N\beta_N} = e^{i\Omega t(\beta_N - \alpha_N)} \sum_{\{\alpha_n, \beta_n\}} \prod_{n=1}^{N} M_{\alpha_n}^{\alpha_n-1} M_{\beta_n}^{\beta_n-1*} \times \prod_{n'=1}^{n} e^{S_{nn'}} \bar{\rho}_{\alpha_0\beta_0}(0) \]
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\]
Augmented Density Matrix & Paths

\[ \overline{\rho}_{\alpha_N \beta_N} = e^{i\Omega t(\beta_N - \alpha_N)} \sum_{\{\alpha_n, \beta_n\}} \prod_{n=1}^{N} M_{\alpha_n}^{\alpha_{n-1}} M_{\beta_n}^{\beta_{n-1} \ast} \times \prod_{n' = n - n_c}^{n} e^{S_{nn'}} \overline{\rho}_{\alpha_0 \beta_0}(0) \]

- Introduce **augmented density matrix** \( R \):

  \( (N. \text{ Makri and D. Makarov, J. Chem. Phys. 102, 4600 (1995)}) \)

  \[ R_n = T_{n, \ldots, n-n_c-1} R_{n-1} \quad \text{Recurrence without memory} \]

- 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

  \( (\alpha_n, \ldots, \alpha_{n-n_c}, \beta_n, \ldots, \beta_{n-n_c}) \)

\[ \Rightarrow \text{Each path is a sequence of the length } 2(n_c + 1) \]

\[ \Rightarrow \text{For a L-level system, there are } L^{2(n_c+1)} \text{ possible paths} \]
Memory depth is given by $n_c \varepsilon$ and has to be chosen sufficiently long:

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

![Graph showing temperature dependence of $\rho_{01}$ for different $\tau_m$.](graph.png)

Long memory depths for
- low temperatures and
- weak fields

M. Glässl et al., PRB 84, 195311 (2011)
Comparison with exact long time asymptotics yields the constraint:

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

Example:

Time dependence of the optical polarization after an ultrafast pulse

- $P(t)/P(0)$
- Time (ps)

$T = 0$ K
$T = 10$ K
$T = 50$ K

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)
Specifics due to the superohmic Coupling

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

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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 $\langle c^\dagger c \, b_q \rangle$
- 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 $\langle c^\dagger c \, b_q b_{q'} \rangle$
- Factorize higher order assisted density matrices like $\langle c^\dagger c \, b_q b_{q'} b_{q''} \rangle$

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


Damped oscillations with renormalized period

Excellent agreement with theoretical predictions!

J. Förstner et al., PRL 91, 127401 (2003)
A. Vagov et al., PRL 98, 227403 (2007)
What characterizes the stationary nonequilibrium state?
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:

\[ H = \hbar \omega_x |X\rangle \langle X| - \hbar g (a^\dagger |0\rangle \langle X| + a |X\rangle \langle 0|) + \sum_q \hbar \omega_q b_q^\dagger b_q + \sum_q \hbar (\gamma_q b_q + \gamma_q b_q^\dagger) |X\rangle \langle X| \]
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Initial cavity preparation:
coherent state with \( \langle n \rangle = 5 \)

\[ g = 0.1 \text{ ps}^{-1} \]
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![Graph showing ground state occupation over time for different temperatures and interaction strengths.](attachment:image.png)
Quantized Light Fields: Revival Dynamics

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Initial cavity preparation:

coherent state with \( \langle n \rangle = 5 \)

Acoustic phonon coupling strongly affects the dynamics, even at \( T = 0 \).
Initial cavity preparation: coherent state with $\langle n \rangle = 5$

![Graph showing ground state occupation over time for different coupling strengths. The graph has a y-axis labeled "ground state occupation" and an x-axis labeled "$g t". The graph compares two cases: the JC-model with $g = 0.1 \text{ ps}^{-1}$ and another case labeled "T = 10 K". The graph shows oscillations in ground state occupation as a function of $g t". The JC-model line is distinct from the "T = 10 K" line, indicating different behaviors for the two cases.]}
Initial cavity preparation: coherent state with $\langle n \rangle = 5$
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)
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

\[ E_0 + \hbar \Omega \rightarrow E_0 + 2\hbar \Omega \]

\[ \sigma^+ \rightarrow \sigma^- \]

\[ |G\rangle \quad |\pm\rangle \quad |B\rangle \]

Polarization parameter

\[ \gamma = \frac{\sigma^+}{\sigma^-} \]

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
Exciton-Biexciton System: Relaxation Dynamics

Energy level sketch

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

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M. Glässl et al., PRB 85, 195306 (2012), M. Glässl et al., to be published
Excitation with a linearly polarized frequency-swept Gaussian pulse (chirp $\alpha$):

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

$$\tau = \sqrt{\frac{\alpha^2}{\tau_0^2} + \tau_0^2}$$
Robust Biexciton Preparation via Adiabatic Rapid Passage

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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