Quantum theory of light-matter interaction in dissipative and non-equilibrium environments

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Outline

1. Light-matter interaction: Some applications
2. Photon-statistics to distinguish light sources
3. Microscopic equation of motion approach
4. Theory and experiment: Unravel underlying physics
5. Theoretical proposals
6. Outlook
Scientific applications

Optical lattices:
Quantum simulators, Quantum algorithms


Generating new states of matter:
Condensates, Polaritons, Superfluids

Plasmonics and subwavelength fabrication


Optomechanics:
Sensing and dissipation studies


Electrooptics.com/

Spectroscopy
Imaging

Transform optics, Metamaterials
New types of light

Consider a beam of photons:

Number of photons within the beam segments (regularity within the beam) determines the statistics.

- random
- bunched
- regular

Physics Today (2009)
Measure the photon statistics

Hanbury Brown-Twiss setup

\[ g^{(2)} = \frac{\langle E^- E^- E^+ E^+ \rangle}{\langle E^- E^+ \rangle^2} \]

unified description requires a fully quantum optical formulation

\[ E(r, t) = \sum_q \left[ \epsilon_q^*(r) c_q^\dagger(t) + \epsilon_q(r) c_q(t) \right] \]

\[ c^\dagger |n\rangle = \sqrt{n+1}|n\rangle \]

\[ c |n\rangle = \sqrt{n}|n-1\rangle \]
Quantify the light statistics

Intensity-intensity correlation

\[ g^{(2)}(t, \tau = 0) = \frac{\langle c^\dagger c^\dagger cc \rangle}{\langle c^\dagger c \rangle^2} \]

random \hspace{1cm} \begin{array}{cccccccccccccccccccccccc} 
\cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \\
\end{array} \hspace{1cm} g^{(2)} = 1

bunched \hspace{1cm} \begin{array}{cccccccccccc} 
\cdots & \cdots & \cdots & \cdots & \cdots & \cdots \end{array} \hspace{1cm} g^{(2)} > 1

regular \hspace{1cm} \begin{array}{cccccccccccccccccccccccc} 
\cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \\
\end{array} \hspace{1cm} g^{(2)} < 1

\[ p_n = \frac{1}{n!} \left( \langle c^\dagger n c^n \rangle - \sum_{j=1}^{N} \frac{(n+j)!}{j!} p_{n+j} \right) \]
Microscopical equations of motion approach
Microscopic Hamiltonian

\[ H = H_{\text{el}} + H_{\text{el-light}} + H_{\text{el-phonon}} \]

- Electronic structure
  - continuum
  - confined states (few level systems)

\[
H_{\text{el}} = \hbar \sum_{m} \omega_{m} \left| m \right\rangle \left\langle m \right| + H_{\text{el-el}}
\]
Microscopic Hamiltonian

\[ H = H_{\text{el}} + H_{\text{el-light}} + H_{\text{el-phonon}} \]

Electronic structure
- continuum
- confined states (few level systems)

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\[ H_{\text{el-light}} = \hbar \sum_k \omega_k c_k^\dagger c_k + \hbar \sum_{m,n} \left( \sum_k M_k^{mn} c_k^\dagger + \Omega_n^m(t) \right) |m\rangle \langle n| + \text{H.c.} \]

Light-matter interaction
- dipole strength/selection rules
- photon states, polarization, angle
Microscopic Hamiltonian

\[ H = H_{el} + H_{el-light} + H_{el-phonon} \]

**Electronic structure**
- continuum
- confined states (few level systems)

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**Light-matter interaction**
- dipole strength/selection rules
- photon states, polarization, angle

\[ H_{el-light} = \hbar \sum_{k} \omega_{k} c_{k}^{\dagger} c_{k} + \hbar \sum_{m,n} \left( \sum_{k} M_{k}^{mn} c_{k}^{\dagger} + \Omega_{n}^{m}(t) \right) |m\rangle \langle n| + H.c. \]

**Electron-phonon interaction**
- longitudinal optical, acoustical phonon
- diagonal coupling, THz frequencies
Strong deviations from Lorentz peaks

\[ g_{L\lambda, q}^{\mu, 3D} = \delta_{\lambda, \mu} \sqrt{\frac{\hbar q}{2\rho c_s}} D_\lambda \]

Acoustic deformation potential

Coupling elements input from material theory
Fröhlich potential

\[ S_{\text{LO},q}^{\lambda \mu,3D} = \frac{1}{q} \sqrt{\frac{e_0^2 \hbar \omega_{\text{LO}}}{2 \varepsilon_0 V}} \left( \frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_\text{st}} \right) \]

1PRB 83, 041304(R) (2011)
Observable dynamics

Heisenberg picture:
\[ \frac{d}{dt} \hat{O} = \frac{i}{\hbar} [H, \hat{O}] \]

Input: microscopic Hamiltonian
\[ H = H_{el} + H_{el-light} + H_{el-phonon} \]

Example: Phonon cQED

\[ G_{n,m}^{p,s} := |g\rangle \langle g| c^\dagger_p c^s b^\dagger_m b^n \]
\[ T_{n,m}^{p,s} := |g\rangle \langle e| c^\dagger_p c^s b^\dagger_m b^n \]
\[ E_{n,m}^{p,s} := |e\rangle \langle e| c^\dagger_p c^s b^\dagger_m b^n \]

Hierarchy problem occurs
Solve with inductive equation of motion approach

All couplings to higher-order photon-phonon correlations included

\[
\frac{d}{dt} G_{n,m}^{p,s} = - [i(m - n)\omega_{LO} + i(p - s)\omega_k] G_{n,m}^{p,s} + iM T_{n,m}^{p+1,s} - iM (T_{m,n}^{s+1,p})^* + isMT_{n,m}^{p,s-1} - ipM^* (T_{m,n}^{s,p-1})^* + ing_v G_{n-1,m}^{p,s} - img_v G_{n,m}^{p,s}
\]

\[
\frac{d}{dt} T_{n,m}^{p,s} = -i [\omega_{eg} - (p - s)\omega_k + (m - n)\omega_{LO}] T_{n,m}^{p,s} - iT_{n,m+1}^{p,s} - iT_{n+1,m}^{p,s} - iM^* (pE_{n,m}^{p-1,s} + E_{n,m}^{p,s+1} - G_{n,m}^{p,s+1}) + ing_v T_{n-1,m}^{p,s} - img_c T_{n,m-1}^{p,s}
\]

\[
\frac{d}{dt} E_{n,m}^{p,s} = - [i(m - n)\omega_{LO} + i(p - s)\omega_k] E_{n,m}^{p,s} - iM T_{n,m}^{p+1,s} - M (T_{m,n}^{s+1,p})^* + ing_v E_{n-1,m}^{p,s} - img_v E_{n,m-1}^{p,s}
\]

1PRL 104, 156801 (2010)

- numerically exact, fast and controllable
- fully quantized optical Bloch equations
- non-equilibrium phonon-photon dynamics computable

\[\hat{b}_q \rightarrow g_q \rightarrow \hat{b}^\dagger_q\]
\[\hat{c} \rightarrow M \rightarrow \hat{c}^\dagger\]
Solve with inductive equation of motion approach

All couplings to higher-order photon-phonon-electron correlations included

$$\frac{dp_n}{dt} = -2\sqrt{n}N\text{Im}(g\langle n\rangle\langle n - 1|a_{1v}^\dagger a_{1c}\rangle) + 2\sqrt{n + 1}N\text{Im}(g\langle n + 1\rangle\langle n|a_{1v}^\dagger a_{1c}\rangle) - 2n\kappa p_n + 2(n + 1)\kappa p_{n+1}.$$ 

$$\frac{d}{dt}\langle n + 1\rangle\langle n|a_{1v}^\dagger a_{1c}\rangle = -\gamma\langle n + 1\rangle\langle n|a_{1v}^\dagger a_{1c}\rangle - \kappa(2n + 1)\langle n + 1\rangle\langle n|a_{1v}^\dagger a_{1c}\rangle + 2\kappa\sqrt{(n+1)(n+2)}\langle n+2\rangle\langle n + 1|a_{1v}^\dagger a_{1c}\rangle$$

$$- P\langle n + 1\rangle\langle n|a_{1v}^\dagger a_{1c}\rangle + \mathcal{F}.$$  \hspace{2cm} (2)

$$\mathcal{F} = ig^*\sqrt{n + 1}(\langle n + 1\rangle\langle n + 1|a_{1v}^\dagger a_{1v}\rangle - \langle n\rangle\langle n|a_{1c}^\dagger a_{1c}\rangle - \langle n + 1\rangle\langle n + 1|a_{1v}^\dagger a_{1c}^\dagger a_{1c} a_{1v}\rangle + \langle n\rangle\langle n|a_{1v}^\dagger a_{1c}^\dagger a_{1c} a_{1v}\rangle)$$

$$- ig^*\sqrt{n + 1}(N - 1) \times (\langle n + 1\rangle\langle n + 1|a_{1v}^\dagger a_{2v}^\dagger a_{1c} a_{2v}\rangle - \langle n\rangle\langle n|a_{1v}^\dagger a_{2v}^\dagger a_{1c} a_{2v}\rangle)$$

$$+ ig\sqrt{n + 2}(N - 1)\langle n + 2\rangle\langle n|a_{1v}^\dagger a_{2v}^\dagger a_{1c} a_{2c}\rangle - ig\sqrt{n}(N - 1)\langle n + 1\rangle\langle n - 1|a_{1v}^\dagger a_{2v}^\dagger a_{1c} a_{2c}\rangle.$$
Solve with inductive equation of motion approach

All couplings to higher-order photon-phonon-electron correlations included

\[
\partial_t \langle G^\dagger G H^{m,n} V^{p,q} \rangle = i \left[ (m - n) \omega_H^0 + (p - q) \omega_V^0 + i \kappa (m + n + p + q) \right] \langle G^\dagger G H^{m,n} V^{p,q} \rangle \\
- i M \ m \langle X_H^\dagger G H^{m-1,n} V^{p,q} \rangle - i M \ n \langle X_H^\dagger G H^{m,n+1} V^{p,q} \rangle - i M \ p \langle X_V^\dagger G H^{m,n} V^{p-1,q} \rangle \\
- i M \ m \langle X_V^\dagger G H^{m,n} V^{p,q+1} \rangle + i M \ n \langle G^\dagger X_H H^{m,n-1} V^{p,q} \rangle + i M \langle G^\dagger X_H H^{m+1,n} V^{p,q} \rangle \\
+ i M \ q \langle G^\dagger X_V H^{m,n} V^{p,q-1} \rangle + i M \langle G^\dagger X_V H^{m,n} V^{p+1,q} \rangle.
\]

\[
\partial_t \langle B^\dagger B H^{m,n} V^{p,q} \rangle = i \left[ (m - n) \omega_H^0 + (p - q) \omega_V^0 + i \kappa (m + n + p + q) \right] \langle B^\dagger B H^{m,n} V^{p,q} \rangle \\
- i M \ m \langle X_H^\dagger B H^{m-1,n} V^{p,q} \rangle + i M \ n \langle X_V^\dagger B H^{m,n+1} V^{p+1,q} \rangle \\
+ i M \langle B^\dagger X_H H^{m,n+1} V^{p,q} \rangle - i M \langle B^\dagger X_V H^{m,n} V^{p,q+1} \rangle.
\]

\[
\partial_t \langle G^\dagger B H^{m,n} V^{p,q} \rangle = i \left[ (m - n) \omega_H^0 + (p - q) \omega_V^0 - \omega_B + i \kappa (m + n + p + q) \right] \langle G^\dagger B H^{m,n} V^{p,q} \rangle \\
- i M \ m \langle X_H^\dagger B H^{m-1,n} V^{p,q} \rangle - i M \ n \langle X_H^\dagger B H^{m,n+1} V^{p,q} \rangle - i M \langle G^\dagger X_H H^{m,n} V^{p,q+1} \rangle \\
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\]

\(^1\text{PRB 81, 195319 (2010)}\)
Explain underlying physics in experiments
Experiments with QD of different electronic configuration (biexciton shifts) lead to a different photon statistics

Experimental data not explainable with rate equations, two-photon transition crucial:

$$\langle G^\dagger B c^\dagger c^\dagger \rangle$$

full quantum electron and photon kinetics of four-level system

two-photon transitions is resonant in case of vanishing biexciton shift and two-photon emission is enhanced

$^1$PRB 87, 245314 (2013)
Steering photon-statistics

Pump strength dependence

Temperature dependence

Microscopic model, adaptable to experimental situation

- dissipation included via Lindblad formalism (incoherent)

\[
\mathcal{L}\rho := \frac{k}{2} \sum_{i=X,B} \mathcal{L}[c_i]\rho + P \sum_{j=H,V} \mathcal{L}[X^\dagger_j G]\rho + \mathcal{L}[B^\dagger X_j]\rho \\
+ \Gamma_{\text{rad}} \sum_{j=H,V} \mathcal{L}[G^\dagger X_j]\rho + \mathcal{L}[X^\dagger_j B]\rho \\
- \sum_{k=G,X,B} \gamma_k (T_k \rho T_k - \rho),
\]

\(^1\text{PRB 87, 245314 (2013)}\)
Phonon-assisted Mollow triplets and phonon lasing

Julia Kabuß and Andreas Knorr

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Phonon-assisted Mollow triplet

Microscopic derivation of time-resolved Raman signal strong excitation limit (non-perturbative in laser and phonon contributions)

PRB 84, 125324 (2011)
Phonon-assisted Mollow triplet

Microscopic derivation of time-resolved Raman signal strong excitation limit (non-perturbative in laser and phonon contributions)

(a) strong excitation

$\Omega_R \{ \begin{array}{ccc}
\omega_l & \rightarrow & \omega_l \\
\omega_l \pm \Omega_c & \rightarrow & \omega_l \pm \Omega_c \\
\omega_l - \omega_{LO} & \rightarrow & \omega_l - \omega_{LO} \\
\end{array} \}$

Stokes Mollow-triplet  Mollow-triplet  anti-Stokes Mollow-triplet

$\Omega_R \{ \begin{array}{ccc}
\omega_{LO} & \rightarrow & \omega_{LO} \\
\omega_{LO} + \Omega_c & \rightarrow & \omega_{LO} + \Omega_c \\
\omega_{LO} - \omega_l & \rightarrow & \omega_{LO} - \omega_l \\
\end{array} \}$

$^1$PRB 84, 125324 (2011)
Phonon-assisted Mollow triplet: Anti-crossings

Time-resolved spectrum of pulsed laser excitation

Laser field amplitude defines hybridisation strength

Read out phonon coupling strength by spectral means without relying on intensity measurements

1PRB 84, 125324 (2011)
Proposal for phonon laser

Use Stokes process in acoustic cavity

- Phonon emission
- Phonon absorption

Phonon assisted Rabi oscillations

\[
\mathcal{H}(t) = \frac{\hbar \omega_{cv}}{2} \sigma_z + \hbar \omega_{ph} b^\dagger b + \Omega(t) \sigma e^{i \omega_L t} + g \sigma^\dagger \sigma b^\dagger + H.c.
\]

\[
\dot{\rho} = -i[\mathcal{H}(t), \rho] + \mathcal{L}\rho
\]

\Gamma_r \quad \text{closes pump cycle}
\Kappa \quad \text{phonon loss}
\gamma_{pd} \quad \text{pure dephasing}

PRL 109, 54301 (2012)
Proposal for phonon laser

Short timescales

Long timescales

$P(n)$

$\Omega_{\text{eff}}^{-1}$

$\approx \Omega_{\text{eff}}^{-1}$

$\approx \Gamma_r^{-1}$

phonon fluctuations

Fock-phonon due to induced Raman process

Coherent phonons

$1$PRL 109, 54301 (2012)
Nanomechanics Strongly Coupled to a Rydberg Superatom

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Cavity optomechanics

- Radiation pressure Hamiltonian
- Small coupling (less than kHz) for membranes

\[ \hbar \omega_{cav}(x) \hat{a}^\dagger \hat{a} \approx \hbar (\omega_{cav} - G\hat{x}) \hat{a}^\dagger \hat{a} \]

\[ \hat{H}_{\text{int}} = -\hbar g_0 \hat{a}^\dagger \hat{a} (\hat{b} + \hat{b}^\dagger) \]

Aspelmeyer et al, arXiv:1303.0733
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Cavity optomechanics – laser driven

- the cavity is driven by a laser \( \rightarrow \) cavity mode is displaced

\[ \hat{a} = \bar{\alpha} + \delta \hat{a} \]

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$$\hat{H}_{\text{int}} = -\hbar g_0 \hat{a}^\dagger \hat{a} (\hat{b} + \hat{b}^\dagger)$$

Cavity optomechanics – laser driven

- the cavity is driven by a laser → cavity mode is displaced
- Radiation pressure Hamiltonian can be linearized → enhanced coupling

$$\hat{a} = \bar{\alpha} + \delta \hat{a}$$

$$\hat{H}_{\text{int}} = -\hbar g_0 (\bar{\alpha} + \delta \hat{a})^\dagger (\bar{\alpha} + \delta \hat{a})(\hat{b} + \hat{b}^\dagger)$$

Aspelmeyer et al, arXiv:1303.0733
Cavity optomechanics: Accomplishments

- successful ground state cooling
- sensing experiments and studies of dissipation processes

Cooling via hybrid system

- utilizing the toolbox of AMO physics to cool down atomic ensemble
- sympathetic cooling by coupling the center of mass motions to the membrane

Hammerer et al, PRL 103, 063005 (2009)
Camerer et al, PRL 107, 223001
Quantum nonlinearities and cavity optomechanics

Cavity optomechanics: Accomplishments
- successful ground state cooling
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Cavity optomechanics: New challenges
- experiments so far in the linear regime
- nonlinearity necessary to create entanglement – to use optomechanics for quantum information processing
Quantum nonlinearities and cavity optomechanics

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- nonlinearity necessary to create entanglement – to use optomechanics for quantum information processing

Our proposal:
use a Rydberg superatom as the nonlinearity in a hybrid system
Rydberg superatom

Rydberg Superatom as an artificial atom

- An atomic ensemble with a Rydberg state interacts strongly due to the VdW interaction → Rydberg shift

Rydberg Superatom as an artificial atom

- An atomic ensemble with a Rydberg state interacts strongly due to the VdW interaction → Rydberg shift
- Rydberg shift leads to the Rydberg blockade mechanism
- Coupling to the light field is increased by the collective enhancement factor

Nanomechanics coupled to a superatom

Nanomechanics Coupled to a Nonlinearity: Hybrid system realization

- use a Rydberg superatom as two-level system
- collective enhancement allows for strong coupling
- Superatom can be pumped, quenched, and can easily be read out
Nanomechanics coupled to a superatom

Principle setup without dissipation processes

\[ H_{\text{int}} = G \left( a^\dagger b + b^\dagger a \right) \]
Nanomechanics coupled to a superatom

\[ H_{\text{int}} = G \left( a^\dagger b + b^\dagger a \right) + \sum_{i=1}^{N} (g_i a^\dagger |e_i\rangle \langle g_i|). \]
Nanomechanics coupled to a superatom

\[ H_{\text{int}} = G \left( a^\dagger b + b^\dagger a \right) + \sum_{i=1}^{N} \left( g_i a |e_i\rangle \langle g_i| + \Omega e^{-i\omega_L t} |r_i\rangle \langle e_i| \right) + \text{h.c.} \]

\[ + \sum_{i,j=1 \atop j > i}^{N} \Delta_R^{ij} |r_i r_j\rangle \langle r_i r_j| + \text{h.c.} \]
Nanomechanics coupled to a superatom: dissipation

Cavity – mediated membrane – Rydberg superatom coupling

- Major obstacles: Dissipation during the excitation transfer
- Phonon decoherence and radiative decay from Rydberg state few kHz
- But: photon leakage and radiative decay from intermediate state MHz
Quantum Monte Carlo Simulation for an ensemble of N=10 three-level atoms
Nanomechanics strongly coupled to superatom

Strong coupling limit is accessible:

\[ G_{\text{eff}} \approx \sqrt{N} \frac{gG\Omega}{\Delta_e \Delta_c} \gg \Gamma_m^{\text{eff}}, \Gamma_r^{\text{eff}}, \Gamma_r, \gamma_m(N_m + 1) \]

\[ \gamma_m \Gamma_m^{\text{eff}} \approx n\kappa \frac{G}{\Delta_c^2} \]

\[ \Gamma_r \approx \Gamma_e \frac{\Omega}{\Delta_e^2} \]

The cavity loss and radiative decay of the intermediate state are suppressed and an effective two-level dynamics take place.

Nanomechanics driven to non-classical state

preparation of non-classical states even at finite temperatures.

Fidelity for the individual state transfer:

\[ F \approx 1 - \frac{\pi}{2G_{\text{eff}}} \left( 4N_m \gamma_m + \gamma_m + \Gamma_r^{\text{eff}} + \Gamma_r \right) \]
What's next
Outlook

- Rydberg physics: transitions from quantum mechanical to classical regimes - quantum path resonances

![Graph showing frequency vs. blockade radius for N=5 atoms]
Outlook

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- Condensed matter physics: disorder in spin chains and disorder induced phase transitions into many-body localization
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- Investigate non-invasive quantum control schemes based on structured continua and develop unified operator technique for quantum feedback
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- Study impact of coherent phonon waves on electronic transport properties (current), cooling, and heating of solids
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Thanks for the attention!