Quantum Electrodynamical TDDFT: From basic theorems to approximate functionals

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TDDFT workshop – Benasque, 2016

Outline

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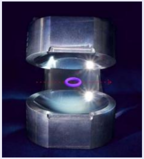
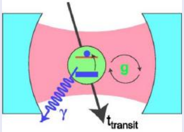
Cavity and circuit QED: Beginning of the story

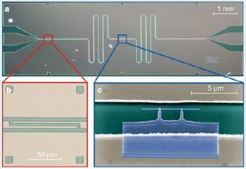
I. Cavity QED (1980s – 1990s): Rydberg atoms in optical cavities



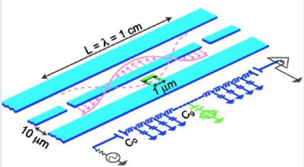


II. Circuit QED (2004): SC qubits in transmission line resonators

“Cooper pair box” in a microwave resonator



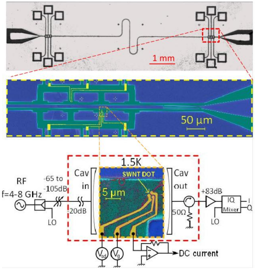
Nature **431**, 162 (2004)



Circuit QED: New generation of quantum circuits

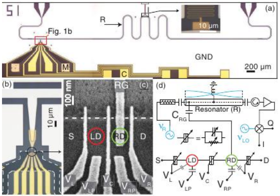
III. Quantum dot strictures integrated with resonators (2011-2012)

SWNTs-based QDs



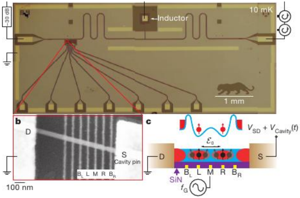
PRL **107**, 256804 (2011)

2DEG-based DQDs



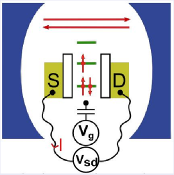
PRL **108**, 046807 (2012)

InAs nanowire-based DQDs



Nature **490**, 380 (2012)

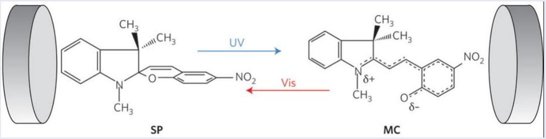
Possibility to experimentally study many-body physics and transport in interacting systems strongly coupled to cavity photons



Cavity QED: “Chemistry-in-cavity”

IV. Molecules in optical cavities (2012)

“Modifying Chemical Landscapes by Coupling to Vacuum Fields”



Angew. Chem. Int. Ed. **51**, 1592 (2012); Nature Mat. **11**, 272 (2012)

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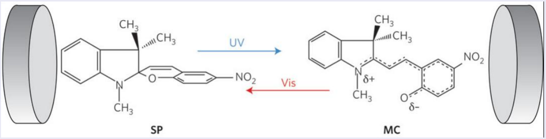


Angew. Chem. Int. Ed. **51**, 1592 (2012); Nature Mat. **11**, 272 (2012)

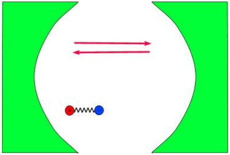
QED-TDDFT: IVT, Phys. Rev. Lett. **110**, 233001 (2013)

- Generalization of TDDFT for the first principle description of non-relativistic many-electron systems interacting with (or driven by) quantum electro-magnetic fields

Outline

Getting started: Classical dipole in classical cavity

Relative coordinate: $\mathbf{x} = \mathbf{x}_+ - \mathbf{x}_-$



Newton equation for dipole dynamics

$$\ddot{\mathbf{x}}(t) = -\Omega^2 \mathbf{x}(t) + \frac{e}{m}\mathbf{E}^{\perp}(\mathbf{r}_0, t)$$

Density of electric current:

$$\mathbf{j}(\mathbf{r}, t) = e\dot{\mathbf{x}}(t)\delta(\mathbf{r} - \mathbf{r}_0) = \partial_t \mathbf{P}(\mathbf{r}, t)$$

Maxwell equations for transverse electromagnetic field, $\nabla \cdot \mathbf{E}^{\perp} = 0$

$$\partial_t \mathbf{B} = -c\nabla \times \mathbf{E}^{\perp}$$

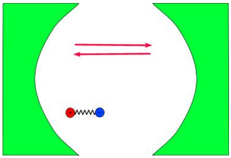
$$\partial_t \mathbf{E}^{\perp} = c\nabla \times \mathbf{B} + 4\pi(\mathbf{j}^{\perp} + \mathbf{j}^{\perp}_{\mathrm{ext}})$$

Transverse part of the current $\mathbf{j}^{\perp} = \partial_t \mathbf{P}^{\perp}$ couples to the cavity modes

$$\mathbf{P}^{\perp}(\mathbf{r}, t) = e\mathbf{x}(t)\delta^{\perp}(\mathbf{r} - \mathbf{r}_0) = \frac{e}{4\pi}\nabla \times \left(\nabla \times \frac{\mathbf{x}(t)}{|\mathbf{r} - \mathbf{r}_0|}\right)$$

Classical cavity (II): the Hamiltonian structure

Electric displacement field: $\mathbf{D}^{\perp} = \mathbf{E}^{\perp} + 4\pi\mathbf{P}^{\perp}$



Maxwell equations in the Hamiltonian form

$$\partial_t \mathbf{B} = -c\nabla \times (\mathbf{D}^{\perp} - 4\pi\mathbf{P}^{\perp}) \mapsto i[H_{\text{e-m}}, \mathbf{B}]$$

$$\partial_t \mathbf{D}^{\perp} = c\nabla \times \mathbf{B} \mapsto i[H_{\text{e-m}}, \mathbf{D}^{\perp}]$$

$$H_{\text{e-m}} = \frac{1}{8\pi}\int d\mathbf{r}\{\mathbf{E}_{\perp}^2 + \mathbf{B}^2\} = \frac{1}{8\pi}\int d\mathbf{r}\{(\mathbf{D}^{\perp} - 4\pi\mathbf{P}^{\perp})^2 + \mathbf{B}^2\}$$

$$[B_i(\mathbf{r}), D_j^{\perp}(\mathbf{r}')] = -i\varepsilon_{ijk}\partial_k\delta(\mathbf{r} - \mathbf{r}')$$

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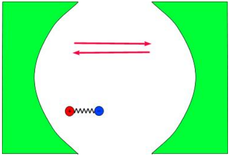

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$$[B_i(\mathbf{r}), D_j^{\perp}(\mathbf{r}')] = -i\varepsilon_{ijk}\partial_k\delta(\mathbf{r} - \mathbf{r}')$$

Projection on cavity modes $\mathbf{E}_\alpha(\mathbf{r})$: $-c^2\nabla^2\mathbf{E}_\alpha = \omega_\alpha^2\mathbf{E}_\alpha\,,\ \nabla\cdot\mathbf{E}_\alpha = 0$

$$\mathbf{D}^{\perp} = \sqrt{4\pi}\sum_\alpha q_\alpha\omega_\alpha\mathbf{E}_\alpha,\ \mathbf{B} = \sqrt{4\pi}\sum_\alpha p_\alpha\frac{c}{\omega_\alpha}\nabla\times\mathbf{E}_\alpha \ \Rightarrow [q_\alpha, p_\beta] = i\delta_{\alpha\beta}$$

$$H_{\text{e-m}} = \frac{1}{2}\sum_\alpha\left[\omega_\alpha^2\left(q_\alpha - \frac{\boldsymbol{\lambda}_\alpha}{\omega_\alpha}\mathbf{x}\right)^2 + p_\alpha^2\right], \quad \boldsymbol{\lambda}_\alpha = e\sqrt{4\pi}\mathbf{E}_\alpha(\mathbf{r}_0)$$

Quantum many-electron system in quantum cavity

Many-body electron-photon wave function

$$\Psi(\{\mathbf{x}_j\},\{q_\alpha\},t)$$

N electrons $\mapsto \{\mathbf{x}_j\}_{j=1}^N$ and M photon modes $\mapsto \{q_\alpha\}_{\alpha=1}^M$

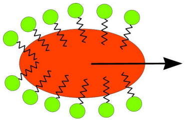
Hamiltonian in a “multipolar” (PWZ) gauge

$$\hat{H} = \sum_j \left[-\frac{1}{2m}\nabla_j^2 + V_{\rm ext}(\mathbf{x}_j,t) \right] + \sum_{i>j} W_{\mathbf{x}_i - \mathbf{x}_j}$$
$$+ \sum_\alpha \left[-\frac{1}{2}\partial_{q_\alpha}^2 + \frac{1}{2}\omega_\alpha^2 \left(q_\alpha - \frac{\boldsymbol{\lambda}_\alpha}{\omega_\alpha}\hat{\mathbf{X}} - \frac{1}{\omega_\alpha} P_{\rm ext}^\alpha \right)^2 \right]$$

$\hat{\mathbf{X}} = \sum_{j=1}^N \mathbf{x}_j$ is the c. m. coordinate of the electronic subsystem

$$i\partial_t \Psi(\{\mathbf{x}_j\},\{q_\alpha\},t) = \hat{H}\Psi(\{\mathbf{x}_j\},\{q_\alpha\},t)$$

Many-electron system harmonically coupled to a set of harmonic oscillators (cavity photons)



Photon induced electron-electron interaction

$$\frac{1}{2}\omega_\alpha^2\left(q_\alpha - \frac{\boldsymbol{\lambda}_\alpha}{\omega_\alpha}\hat{\mathbf{X}}\right)^2 \mapsto -\underbrace{\sum_j \omega_\alpha \hat{q}_\alpha \boldsymbol{\lambda}_\alpha \hat{\mathbf{x}}_j}_{\sim(\hat{a}^\dagger+\hat{a})\psi^\dagger\psi} + \underbrace{\frac{1}{2}\sum_{i,j}(\boldsymbol{\lambda}_\alpha \hat{\mathbf{x}}_i)(\boldsymbol{\lambda}_\alpha \hat{\mathbf{x}}_j)}_{\sim v(\mathbf{x},\mathbf{x}')\hat{n}(\mathbf{x})\hat{n}(\mathbf{x}')}$$

Effective interaction

$$\mathcal{W}_{\text{eff}}^{\alpha}(\mathbf{x},t;\mathbf{x}',t') = \boldsymbol{\lambda}_\alpha \hat{\mathbf{x}} \underbrace{\left[\omega_\alpha^2 \langle \hat{q}_\alpha(t)\hat{q}_\alpha(t')\rangle + \delta(t-t')\right]}_{\mathcal{D}(t-t')} \boldsymbol{\lambda}_\alpha \hat{\mathbf{x}}'$$

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$$\mathcal{D}(\omega) = \underbrace{\frac{\omega_\alpha^2}{\omega^2-\omega_\alpha^2}}_{\langle \mathbf{D}_\perp\cdot\mathbf{D}_\perp\rangle_\omega} + 1 = \underbrace{\frac{\omega^2}{\omega^2-\omega_\alpha^2}}_{\langle \mathbf{E}_\perp\cdot\mathbf{E}_\perp\rangle_\omega} \mapsto \ddot{\mathbf{x}}$$

Physical electron-electron interaction is mediated by the electric field.

Only accelerated electron generates the transverse electric field felt by another electron!

Outline

Direct map: $\{\Psi_0, V_{\text{ext}}, P^{\alpha}_{\text{ext}}\} \mapsto \{\Psi, n, Q_\alpha\}$

$$i\partial_t \Psi(\{\mathbf{x}_j\},\{q_\alpha\},t) = \hat{H}\Psi(\{\mathbf{x}_j\},\{q_\alpha\},t); \quad \Psi(t=0)=\Psi_0$$

$$\hat{H} = \sum_j \left[-\frac{\nabla_j^2}{2m} + V_{\text{ext}}(\mathbf{x}_j,t)\right] + \sum_{i>j} W_{\mathbf{x}_i-\mathbf{x}_j}$$
$$+\sum_\alpha \Bigg[\underbrace{-\frac{1}{2}\partial^2_{q_\alpha}}_{\mathbf{B}^2} + \underbrace{\frac{\omega_\alpha^2}{2}\left(q_\alpha - \frac{\boldsymbol{\lambda}_\alpha}{\omega_\alpha}\hat{\mathbf{X}} - \frac{1}{\omega_\alpha}P^{\alpha}_{\text{ext}}\right)^2}_{\mathbf{E}^2_\perp}\Bigg]$$

Basic variables: mean photon coodinates and the electronic density

$$Q_\alpha(t) = \langle\Psi(t)|q_\alpha|\Psi(t)\rangle \sim \text{ electric displacement}$$
$$n(\mathbf{x},t) = \langle\Psi(t)|\hat{n}(\mathbf{x})|\Psi(t)\rangle$$

$$\hat{\mathbf{X}} = \int \mathbf{x}\hat{n}(\mathbf{x})d\mathbf{x}, \qquad \mathbf{R}(t) = \langle\Psi(t)|\hat{\mathbf{X}}|\Psi(t)\rangle = \int \mathbf{x}n(\mathbf{x},t)d\mathbf{x}$$

Inverse map: $\{\Psi_0, n, Q_\alpha\} \mapsto \{\Psi, V_{\rm ext}, P^\alpha_{\rm ext}\}$ via NLSE

Starting point: Equations of motion for basic variables

"Maxwell": $\ddot{Q}_\alpha + \omega_\alpha^2 Q_\alpha - \omega_\alpha \boldsymbol{\lambda}_\alpha \mathbf{R} = \omega_\alpha P^\alpha_{\rm ext}$,

"Force balance": $m\ddot{n} + \nabla \mathbf{F}_{\rm str} + \sum_\alpha \nabla \mathbf{f}_\alpha = \nabla\left[n(\nabla V_{\rm ext} + \boldsymbol{\lambda}_\alpha P^\alpha_{\rm ext})\right]$,

$$\mathbf{F}_{\rm str}(\mathbf{x},t) = im\langle\Psi|[\hat{T}+\hat{W},\hat{j}_p]|\Psi\rangle = -\nabla \overleftrightarrow{\Pi}$$ stress force

$$\mathbf{f}_\alpha(\mathbf{x},t) = \boldsymbol{\lambda}_\alpha\langle\Psi|(\omega_\alpha q_\alpha - \boldsymbol{\lambda}_\alpha \hat{\mathbf{X}})\hat{n}(\mathbf{x})|\Psi\rangle$$ force from α-mode

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$V_{\rm ext}[n,\Psi],\ P^\alpha_{\rm ext}[Q_\alpha, n]:\ \hat{H}[V_{\rm ext}, P^\alpha_{\rm ext}] \mapsto \hat{H}[n, Q_\alpha, \Psi] \implies$ NLSE

This NLSE defines the TDDFT map: $\{\Psi_0, n, Q_\alpha\} \mapsto \{\Psi, V_{\rm ext}, P^\alpha_{\rm ext}\}$

QED-TDDFT mapping theorem

$|\Psi(t)\rangle$, $V_{\rm ext}(\mathbf{x},t)$, and $P^\alpha_{\rm ext}(t)$ are unique functionals of the initial state $|\Psi_0\rangle$ and the basic observables $n(\mathbf{x},t)$, and $Q_\alpha(t)$.

Comments on mathematical issues

Nonlinear many-body problem for QED-TDDFT

$$i\partial_t|\Psi\rangle = \hat{H}[V_{\text{ext}}, P_{\text{ext}}^{\alpha}]|\Psi\rangle, \quad |\Psi(t=0)\rangle = |\Psi_0\rangle$$

$$\ddot{Q}_\alpha + \omega_\alpha^2 Q_\alpha - \omega_\alpha \boldsymbol{\lambda}_\alpha \mathbf{R} = \omega_\alpha P_{\text{ext}}^{\alpha}$$

$$m\ddot{n} + \nabla \mathbf{F}_{\text{str}}[\Psi] + \sum_\alpha \nabla \mathbf{f}_\alpha[\Psi] = \nabla\left[n(\nabla V_{\text{ext}} + \boldsymbol{\lambda}_\alpha P_{\text{ext}}^{\alpha})\right]$$

Solve for $|\Psi(t)\rangle$, $V_{\text{ext}}(\mathbf{x}, t)$, and $P_{\text{ext}}^{\alpha}(t)$, given $|\Psi_0\rangle$, $n(\mathbf{x}, t)$, and $Q_\alpha(t)$

Existence of a unique solution: QED-TDDFT mapping theorem

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Currently we have (assuming a finite number of photon modes):

- “Standard” proof of uniqueness under the assumption of t-analyticity [IVT, PRL **110**, 233001 (2013)]
- Rigorous proof of uniqueness and existence for lattice electrons [M. Farzanepour and IVT, PRB **90**, 195149 (2014)]

Kohn-Sham construction for the QED-TDDFT

"Maxwell-Schrödinger" dynamics for N noninteracting KS particles

$$i\partial_t \phi_j = -\frac{\nabla^2}{2m}\phi_j + \left[V_S + \sum_\alpha (\omega_\alpha Q_\alpha - \boldsymbol{\lambda}_\alpha \mathbf{R} - P^\alpha_{\text{ext}})\boldsymbol{\lambda}_\alpha \mathbf{x}\right]\phi_j,$$

$$\ddot{Q}_\alpha + \omega_\alpha^2 Q_\alpha - \omega_\alpha(\boldsymbol{\lambda}_\alpha \mathbf{R} + P^\alpha_{\text{ext}}) = 0$$

The KS density reproduces the physical density, $n_S(\mathbf{x},t) = n(\mathbf{x},t)$, if

$$V_S = V_{\text{ext}} + V^{\text{el}}_{\text{Hxc}}[n, Q_\alpha] + \sum_\alpha V^\alpha_{\text{xc}}[n, Q_\alpha]$$

The "electronic" $V^{\text{el}}_{\text{Hxc}}[n, Q_\alpha]$ and "photonic" $V^\alpha_{\text{xc}}[n, Q_\alpha]$ xc potentials

$$\nabla(n\nabla V^{\text{el}}_{\text{Hxc}}) = \nabla(\mathbf{F}^S_{\text{str}} - \mathbf{F}_{\text{str}}) = \nabla(\nabla \overleftrightarrow{\Pi}_{\text{Hxc}}),$$

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Zero force theorem holds true for both xc potentials $V^{\text{el}}_{\text{Hxc}}$ and V^α_{xc}

$$\int n\nabla V^{\text{el}}_{\text{Hxc}} d\mathbf{x} = \int n\nabla V^\alpha_{\text{xc}} d\mathbf{x} = 0$$

The problem of approximations

We succeeded to define xc potentials in the electron-photon TDDFT in such a way, that they have similar general properties and satisfy similar constraints as $V_{\rm xc}$ in the usual purely electronic TDDFT.

Natural approximation strategies

I. "Velocity gradient expansion":

- At "zero level" we set $V_{\rm xc}^{\alpha} = 0$ and take $V_{\rm xc}^{\rm el} = V_{\rm xc}^{\rm ALDA}$. This 0-approximation correctly reproduces a "HPT-type" rigid motion with a uniform velocity – analog of ALDA
- Quantum/nonadiabatic corrections $\sim \nabla \mathbf{v}(\mathbf{x}, t)$ – similar to Vignale-Kohn construction in the electronic TDCDFT

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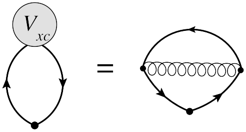
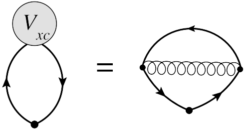
II. "OEP-strategy":

- Making a connection to the many-body theory via electron-photon generalization of the Sham-Schlütter equation.



Electron-photon Optimized Effective Potential[1]



Formulation in terms of orbital xc functionals: $V_{\rm xc} = \delta E_{\rm xc}/\delta n$

Ground state: Interpret Lamb shift as xc orbital functional

$$E_{\rm xc} = \frac{1}{2} \sum_{\alpha,k,j} f_k (1 - f_j) \frac{\epsilon_j - \epsilon_k}{\epsilon_j - \epsilon_k + \omega_\alpha} \langle \phi_j | \boldsymbol{\lambda}_\alpha \mathbf{x} | \phi_k \rangle \langle \phi_k | \boldsymbol{\lambda}_\alpha \mathbf{x} | \phi_j \rangle$$

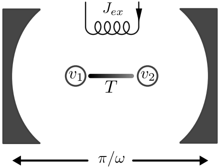
Dynamics: Schwinger-Keldysh xc action functional

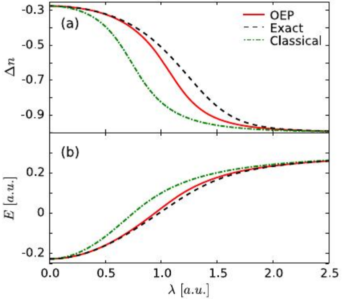
$$A_{\rm xc} = \sum_{\alpha,k,j} \int_{\mathcal{C}} d\tau_1 d\tau_2 f_k (1 - f_j) \mathcal{W}^{>}(\tau_1, \tau_2) \langle \phi_j | \boldsymbol{\lambda}_\alpha \mathbf{x} | \phi_k \rangle |_{\tau_1} \langle \phi_k | \boldsymbol{\lambda}_\alpha \mathbf{x} | \phi_j \rangle |_{\tau_2}$$

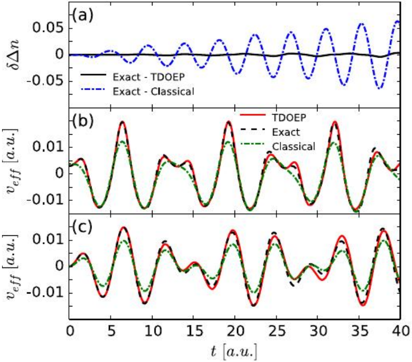
[1] C. Pellegrini, et. al., PRL **115**, 093001 (2015)

Test case: 2-site “molecule” coupled to a singe mode

$$\hat{H} = -T\sigma_x + V_{\text{ext}}(t)\sigma_z - \frac{1}{2}\partial_q^2 + \frac{\omega^2}{2}\left(q - \frac{\lambda}{\omega}\sigma_z\right)^2$$







Outline

Connection to the theory of open quantum systems

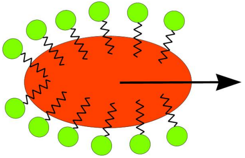
Electron-photon dynamics without a "photon driving field", $P^{\alpha}_{\text{ext}}(t) = 0$

$$i\partial_t \Psi(\{\mathbf{x}_j\},\{q_\alpha\},t) = \hat{H}\Psi(\{\mathbf{x}_j\},\{q_\alpha\},t); \quad \Psi(t=0) = \Psi_0$$

$$\hat{H} = \sum_j \left[-\frac{\nabla_j^2}{2m} + V_{\text{ext}}(\mathbf{x}_j,t) \right] + \sum_{i>j} W_{\mathbf{x}_i - \mathbf{x}_j} + \sum_\alpha \left[-\frac{1}{2}\partial^2_{q_\alpha} + \frac{\omega_\alpha^2}{2}\left(q_\alpha - \frac{\boldsymbol{\lambda}_\alpha}{\omega_\alpha}\hat{\mathbf{X}} \right)^2 \right]$$

Many-electron system harmonically coupled to a set of harmonic oscillators (cavity photons)

Caldeira-Leggett model of dissipative quantum systems [Ann. Phys. **149**, 374 (1983)]



TDDFT for open quantum systems

KS equations in the cavity TDDFT with $P^{\alpha}_{\text{ext}}(t) = 0$

$$i\partial_t \phi_j = -\frac{\nabla^2}{2m}\phi_j + \Big[V_{\text{ext}} + V_{\text{Hxc}}[n, Q_\alpha] + \textstyle\sum_\alpha (\omega_\alpha Q_\alpha - \boldsymbol{\lambda}_\alpha \mathbf{R})\boldsymbol{\lambda}_\alpha \mathbf{x}\Big]\phi_j,$$

$$\ddot{Q}_\alpha + \omega_\alpha^2 Q_\alpha = \omega_\alpha \boldsymbol{\lambda}_\alpha \mathbf{R}(t), \quad \mathbf{R}(t) = \textstyle\int \mathbf{x} n(\mathbf{x}, t) d\mathbf{x}$$

"Tracing out" photons (bath): $Q_\alpha[n](t) = \int_0^t \sin[\omega_\alpha(t - t')]\boldsymbol{\lambda}_\alpha \mathbf{R}(t')dt'$

Closed KS equations for an open quantum system

$$i\partial_t \phi_j = -\frac{\nabla^2}{2m}\phi_j + \big(V_{\text{ext}} + V_{\text{eff}}[n]\big)\phi_j,$$

$$V_{\text{eff}}[n] = V_{\text{Hxc}}[n, Q_\alpha[n]] + \textstyle\sum_\alpha (\omega_\alpha Q_\alpha[n] - \boldsymbol{\lambda}_\alpha \mathbf{R})\boldsymbol{\lambda}_\alpha \mathbf{x}$$

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Ohmic spectral density of the bath $\pi \sum_\alpha \lambda_\alpha^\mu \lambda_\alpha^\nu \delta(\omega - \omega_\alpha) = 2\eta\delta^{\mu\nu} \implies$

$$V_{\mathrm{eff}} = V_{\mathrm{Hxc}} + \eta N \dot{\mathbf{R}}\mathbf{x}$$

In "zero-level" approximation we recover Albrecht's dissipative NLSE [Phys. Lett. B **56**,127 (1975)]

Outline

Summary

- It is possible to formulate a rigorous TDDFT approach to address dynamics of many-electron systems strongly coupled to cavity/resonator photons
- General properties of corresponding xc potentials suggest several strategies for constructing approximations. We constructed and tested an electron-photon QED-OEP functional (work is still in progress)
- QED-TDDFT leads to a very natural formulation of TDDFT for open/dissipative quantum systems:

  (i) first set up TDDFT together with approximation, and
  (ii) then “trace out” the bath
- QED-TDDFT is naturally formulated in the dipole approximation. Beyond dipole approximation coupling to the photon's magnetic field has to be considered. Hence one should use the electron current $\mathbf{j}(\mathbf{x}, t)$ as a basic variable $\Rightarrow$ QED-TDCDFT.

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