Krieger-Li-Iafrate approximation to the optimized effective potential approach in density functional theory for quantum electrodynamics

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Introduction

Many-body perturbation theory (MBPT) opens the possibility to construct approximations to every desired order of a 'weak' interacting system. The drawback is a in general non-local interaction in space and time and it is therefore a demanding task to apply it to 'real' systems.

The optimized effective potential (OEP), derived by inversion of the Sham-Schlüter equation, is a natural connection between local density-functional theory and MBPT. In principle, this variationally best local potential reduces the problem to solving a simple system of Kohn-Sham equations combined with the solution of the OEP integral equation. However, converging the full set of OEP equations is a quite challenging procedure and is in practice rarely tackled.

The Krieger-Li-Iafrate (KLI) approximation reduces the integral equation to an analytically solvable one via a dominant orbital approximation. It performs usually quite well for electronic systems.

In the present work, we extend the OEP and KLI approaches to the case of electron-photon interactions in quantum optics and quantum electrodynamics. Here an effective electronic interaction is transmitted via transversal photons. We present first static and time-dependent results for the OEP [1] and KLI approximations of the Rabi model and compare with the exact configuration-interaction solution and the corresponding exact Kohn-Sham potentials. [2]

References: [1] C. Pellegrini et.al., arXiv 1412.4530 (2014). [2] M. Ruggenthaler et.al., Phys. Rev. A 90, 012508 (2014).

From many-body perturbation theory to density functional theory

One can derive the OEP in terms of the electronic Greens functions on the Keldysh contour.

One particle Greens function on the Keldysh contour →

$$G(1;1') = \frac{1}{i}\frac{\langle\Psi_0|\hat{T}\left(e^{-i\int_\gamma dz\hat{H}(z)}\hat{\psi}(1)\hat{\psi}^\dagger(1')\right)|\Psi_0\rangle}{\langle\Psi_0|\Psi_0\rangle}$$

Kadanoff-Baym equations for interacting and Kohn-Sham Greens functions

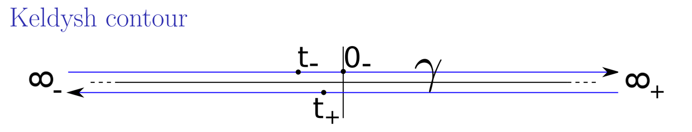
$$\left[i\frac{d}{dz_1} - h(1)\right]G(1;1') = \delta(1;1') \pm i\int d2\Sigma_{xc}(1;2)G(2;1')$$

$$\left[i\frac{d}{dz_1} - h_s(1)\right]G_s(1;1') = \delta(1;1')$$

Gordon Baym and Leo P Kadanoff, Phys. Rev. **124**, 287 (1961).

Assuming an adiabatic switch, therefore neglecting the Matsubara-track, we arrive at the

Keldysh contour



Sham-Schlüter equation

Using the Kubo-Martin-Schwinger boundary conditions and the definition of the Kohn-Sham system $n(1) = -iG^<(1,1^+) = -iG_s^<(1,1^+)$, one arrives at the Sham-Schlüter equation in the following generalized form

$$\int d2\int d3G_s(1,2)\Sigma_{xc}(2,3)G(3,1) = \int d2G(1,2)v_{xc}(2)G_s(2,1)$$

The **solution** of this equation is the **optimized effective potential** (OEP) v_{xc} that is local in time and space and mimics all many-body exchange and correlation effects.

Robert van Leeuwen, Phys. Rev. Lett., **76**(19):3610, (1996).

Hamiltonian - electron-photon coupling in dipole approximation

Hamiltonian

$$\hat{H} + \hat{H}_0 + \frac{1}{2}\sum_\alpha\left[\hat{\boldsymbol{p}}_\alpha^2 + \omega_\alpha^2\left(\hat{\boldsymbol{q}}_\alpha - \frac{\lambda_\alpha}{\omega_\alpha}\hat{\boldsymbol{R}}\right)^2\right]$$

where the electronic interaction is mediated by **transversal** photons.

Interaction

The corresponding second-order interaction is given by

$$W(1;2) = \sum_\alpha \lambda_\alpha\boldsymbol{r}_1\cdot\lambda_\alpha\boldsymbol{r}_2\left(\omega_\alpha^2 D(t_1;t_2) + \delta(t_1-t_2)\right)$$

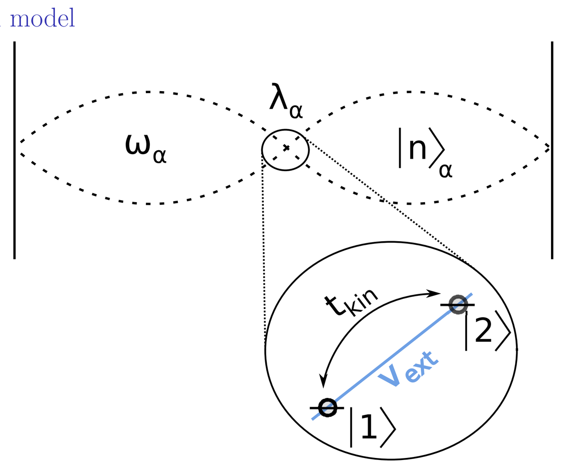
with the transversal photon-propagator $D(t_1;t_2)$.

The corresponding self-energy is approximated by the single-shot-like G_sW_0 approximation, with $T\to 0$

$$\Sigma_x(1;2) = iG_s(1;2)W_0(2;1)$$

C. Pellegrini et.al., arXiv 1412.4530 (2014).

Rabi model



$$\hat{H} = -t_{kin}\hat{\sigma}_x + \left[g_\alpha\left(\hat{a}_\alpha + \hat{a}_\alpha^\dagger\right) + v_{ext}(t)\right]\hat{\sigma}_z + \omega_\alpha\left(\hat{a}_\alpha^\dagger\hat{a}_\alpha + \frac{1}{2}\right) + \frac{\lambda_\alpha^2}{2}$$

with coupling $g_\alpha = \sqrt{\frac{\omega_\alpha}{2}}\lambda_\alpha$ and the Pauli-matrices

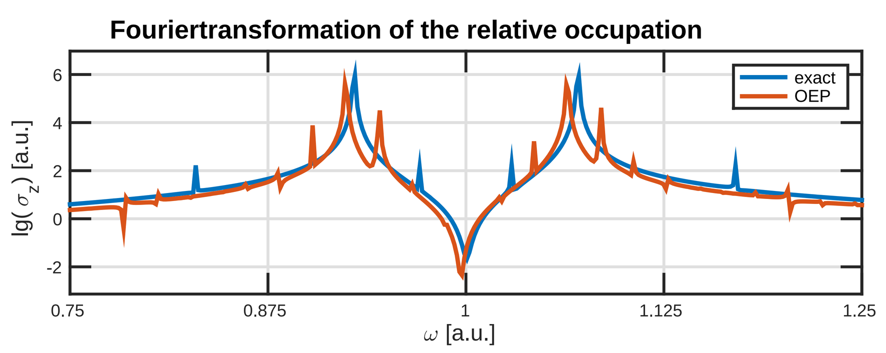
$$\sigma_z = \begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix}\quad \sigma_x = \begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix}$$

OEP over-screening

Comparison of full TDOEP integral equation with TDCI

We encounter a **beat** frequency in the **resonant** Rabi-solution → possible consequence of G_sW_0 **over-screening**.

Improvable via the T-matrix approximation in orders of W_0?



Static Krieger-Li-Iafrate approximation

Static OEP equation in density functional theory for quantum-electrodynamics

$$\sum_{i,j\neq i}\frac{\phi_j^*(\boldsymbol{r})\phi_i(\boldsymbol{r})}{\varepsilon_i-\varepsilon_j}\left(f_i\langle\phi_i|v_x|\phi_j\rangle - \sum_{k,\alpha}\frac{d_{ik}^\alpha d_{kj}^\alpha}{2}(\varepsilon_i-\varepsilon_k)\left[\frac{f_i(1-f_k)}{\varepsilon_i-\varepsilon_k-\omega_\alpha} + \frac{(1-f_i)f_k}{\varepsilon_i-\varepsilon_k+\omega_\alpha}\right]\right) + c.c.$$
$$= \frac{1}{2}\sum_{\alpha,i,k}\omega_\alpha\frac{|d_{ik}^\alpha|^2 f_i(1-f_k)}{(\varepsilon_i-\varepsilon_k-\omega_\alpha)^2}\left(|\phi_k(\boldsymbol{r})|^2 - |\phi_i(\boldsymbol{r})|^2\right)$$

C. Pellegrini et.al., arXiv 1412.4530 (2014).

Converging the above integral equation is quite challenging and we therefore try to isolate the potential from the integral.

Energy denominator approximation

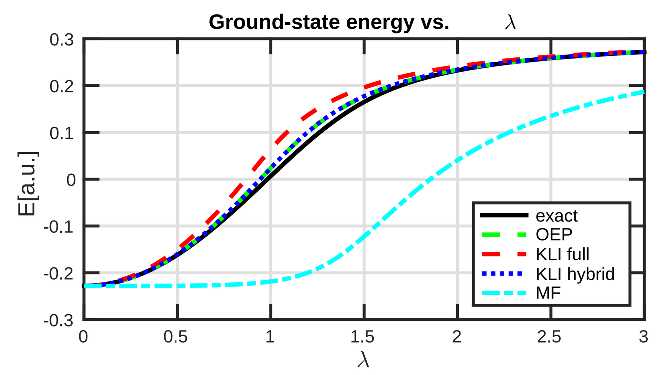
$$\frac{1}{\varepsilon_i-\varepsilon_j} = \frac{1}{\mp\Delta\varepsilon}\quad \mp\quad \text{for}\quad i</>N_{\text{Fermi}}$$

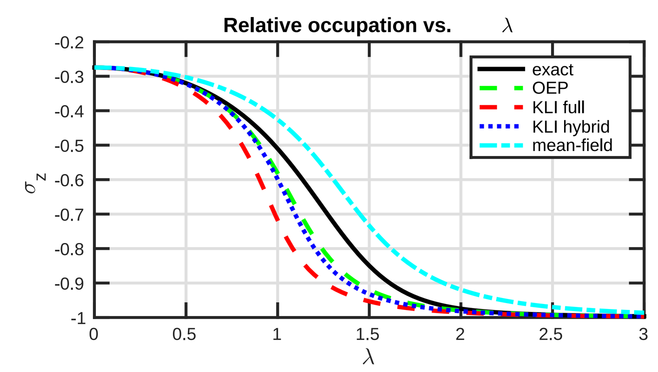
Where $\Delta\varepsilon$ can be interpreted as the dominant orbital or dipole transition energy.

→ **dependence on** $\Delta\varepsilon$, at least through rhs
not the case for Coulomb interaction
→ different levels of approximation possible

- **basic** - just relevant part
- **hybrid** - complete left hand side
- **full** - all energy differences

Observables for the Rabi model





$t_{kin} = 0.7\quad v_{ext} = 0.2\quad \omega_\alpha = 1.0$ with $\Delta\varepsilon_{\text{hybrid}} = \text{static}$

- **basic** → unstable beyond weak coupling
- **hybrid** → performing well with (iterative) dominant dipole
- **full** → performing well with linear response theory

$\Delta\varepsilon$ determination

Linear response - f-sum-rule

Using the f-sum-rule for one electron

$$-\frac{1}{\pi}\int d\omega\omega\Im(\chi_{R,s}(\boldsymbol{r},\boldsymbol{r}',\omega)) = c_1 = N \stackrel{!}{=} 1$$

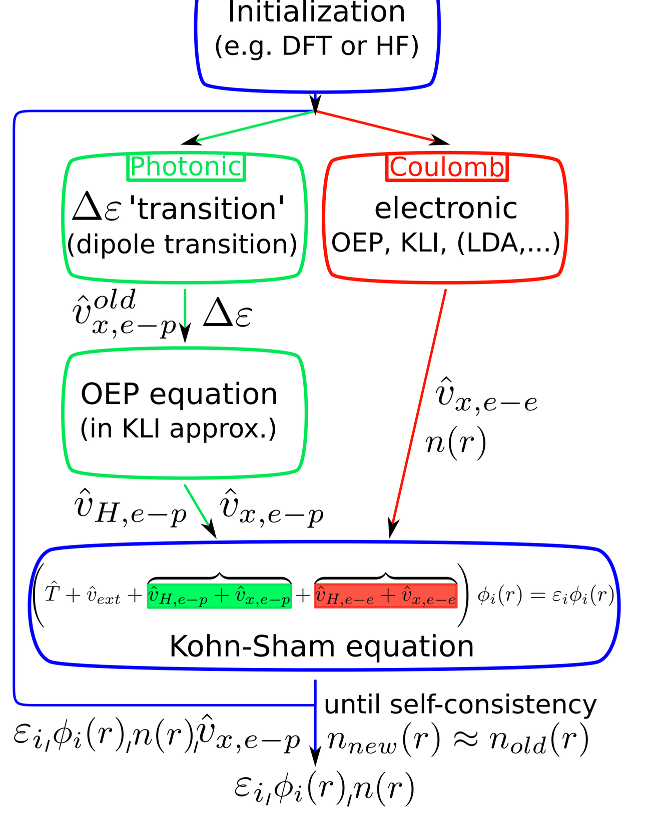
in the Kohn-Sham-representation with

$$\Im\left(\chi_{R,s}(\boldsymbol{r},\boldsymbol{r}',\omega)\right) = -\pi\sum_{k,l}f_{N,k,l}(\boldsymbol{r})f_{N,k,l}(\boldsymbol{r}')\left[\delta(\omega-(\varepsilon_k-\varepsilon_l)) - \delta(\omega+(\varepsilon_k-\varepsilon_l))\right]$$
$$\text{with}\quad f_{N,k,l}(\boldsymbol{r}) = f_l(1-f_k)\phi_l^*(\boldsymbol{r})\phi_k(\boldsymbol{r})$$

and applying the energy denominator approximation

Iterative dominant dipole transition

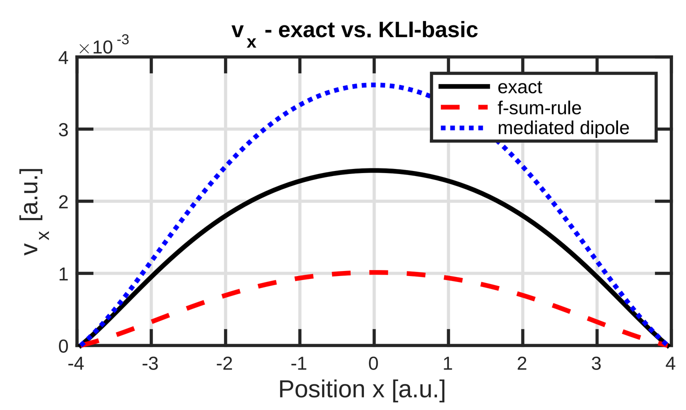
Calculate $\Delta\varepsilon[\overline{d_{ij}}]$ or $\Delta\varepsilon[d_{ij}^{\max}]$ within the self-consistent loop



1D hydrogen atom

Comparison between exact diagonalization and KLI-basic

Implementation still in progress.
First results for KLI-basic in combination with f-sum-rule and different dominant dipole transition methods.
Qualitative agreement for weak, $\lambda_\alpha = 0.01$, coupling →
but unstable beyond weak coupling.
$\omega_\alpha = \varepsilon_2 - \varepsilon_1$



Time-dependent KLI

We follow CA Ullrich, UJ Gossmann, EKU Gross, Phys. Rev. Lett., **74**(6):872, (1995), however the physical motivation is limited, since the static representation of the mean-value approximation to

$$p_j(\boldsymbol{r}t) = \frac{-i}{\phi_j^*(\boldsymbol{r}t)}\int_{-\infty}^{\infty}dt_1\int d\boldsymbol{r}_1\left[f_jv_x(\boldsymbol{r}_1t_1) - u_{x_j}(\boldsymbol{r}_1t_1)\right]\phi_j^*(\boldsymbol{r}_1t_1)\sum_{k\neq j}\phi_k^*(\boldsymbol{r}t)\phi_k(\boldsymbol{r}_1t_1)\theta(t-t_1)$$

holds no longer true. The time-dependent KLI potential is given by

$$v_x^{tdKLI}(\boldsymbol{r}t) = \frac{1}{n(\boldsymbol{r}t)}\sum_j\frac{n_j(\boldsymbol{r}t)}{2}\left[u_{x_j}(\boldsymbol{r}t) + u_{x_j}^*(\boldsymbol{r}t)\right] + \frac{1}{n(\boldsymbol{r}t)}\sum_j n_j(\boldsymbol{r}t)\left[f_j\overline{v}_{x_j}(t) - \frac{1}{2}\left(\overline{u}_{x_j}(t) + \overline{u}_{x_j}^*(t)\right)\right]$$
$$+\frac{i}{4n(\boldsymbol{r}t)}\sum_j\left(\nabla^2 n_j(\boldsymbol{r}t)\right)\int_{-\infty}^{t}dt_1\left[\overline{u}_{x_j}(t_1) - \overline{u}_{x_j}^*(t_1)\right]$$

where just the last part remains for the Rabi-model and $u_{xc_j}(\boldsymbol{r}_1t_1)$ is given by

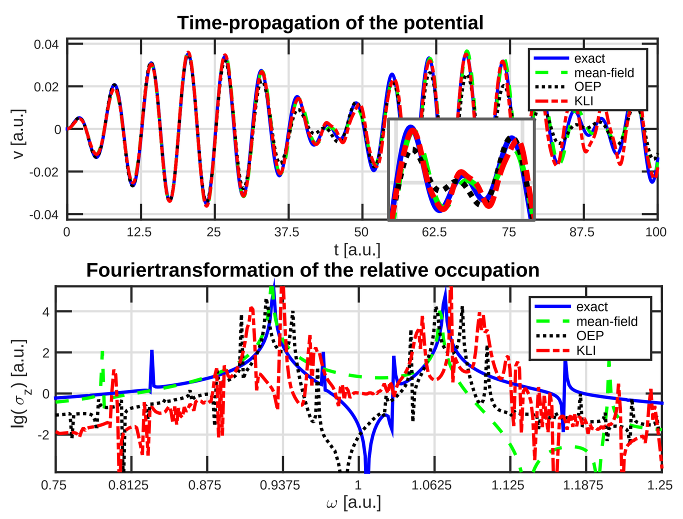
$$u_{x_j}(\boldsymbol{r}_1t_1) = \frac{1}{\phi_j^*(\boldsymbol{r}_1t_1)}\sum_{k,\alpha}\int_{-\infty}^{t_1}dt_2d_{jk}^\alpha(t_2)\left[(1-f_j)f_kW^>(t_1,t_2) - f_j(1-f_k)W^<(t_1,t_2)\right]\boldsymbol{\lambda}_\alpha\hat{\boldsymbol{r}}_1\phi_k^*(\boldsymbol{r}_1t_1)$$

Initial-state

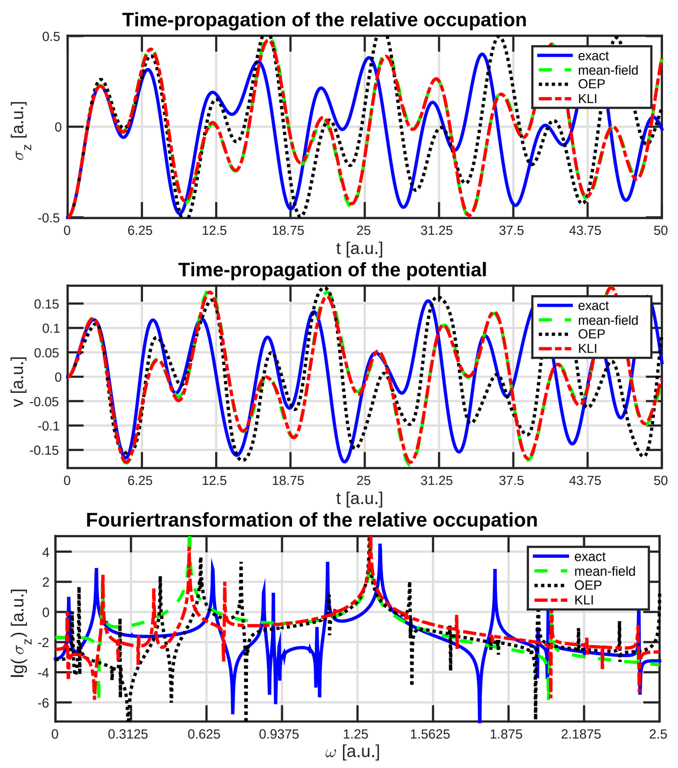
Sudden switch in coupling $\lambda(t) = \lambda\theta(t)$ with the initial-state $|\Psi(t=0)\rangle = \left(\frac{1}{2}|1\rangle + \frac{\sqrt{3}}{2}|2\rangle\right)\otimes|0\rangle$ and an external potential $v_{ext} = 0$.

Resonant

$\lambda = 0.1,\ \omega = 1,\ t_{kin} = 0.5$

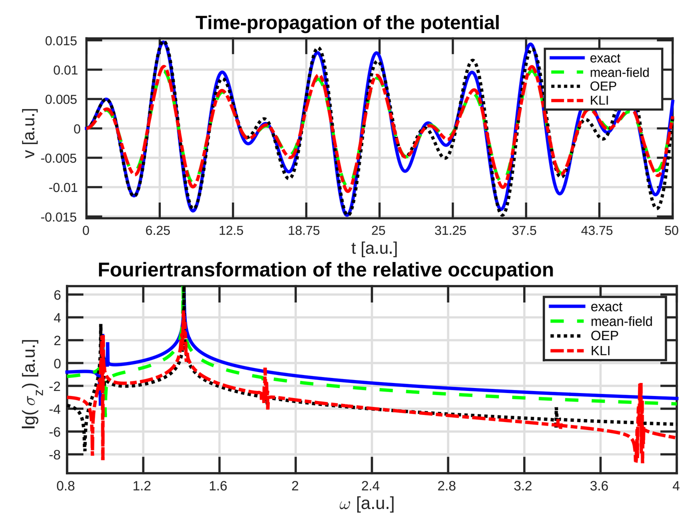


$\lambda = 0.5$

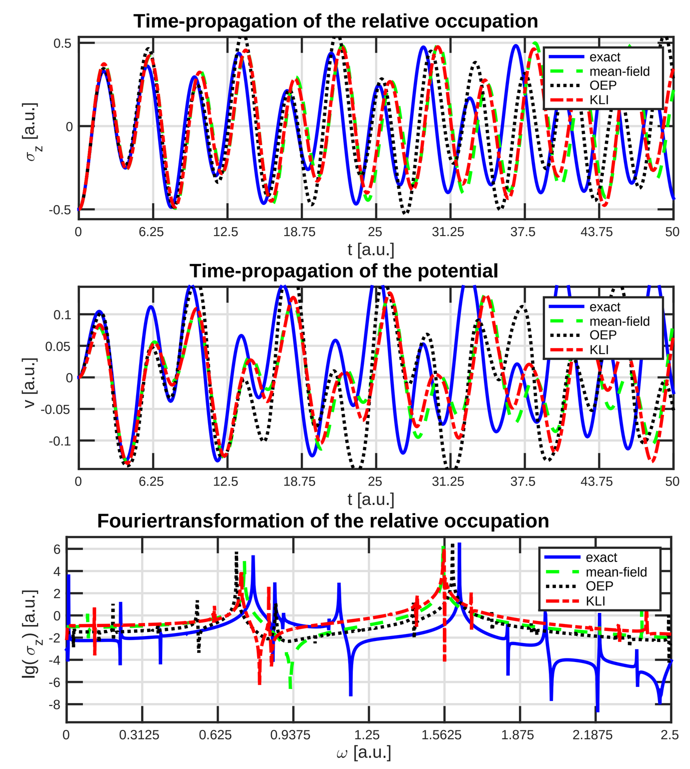


Off-resonant

$\lambda = 0.1,\ \omega = 1,\ t_{kin} = 0.7$



$\lambda = 0.5$



Slight improvement compared to mean-field, especially in the vicinity of nodal points and maxima.

Summary and Outlook

The retarded interaction mediated by transversal photons increases the complexity for good approximations. The remaining dependence on $\Delta\varepsilon$ and a more physical motivation has to be cleared.

The (TD)KLI approximation can be seen as a small correction beyond the mean-field approximation, including a smoother limit $\lambda_\alpha\to\infty$ and a small correction in the vicinity of nodal-points and maxima, where the exchange-part is more relevant.

In future, we plan to apply the TDKLI approximation to a 1D-hydrogen model to check the relevance of the OEP/KLI concept for quantum-electrodynamics beyond simple quantum-optical model systems.