Linear-Response Formulation of TDDFT for QED

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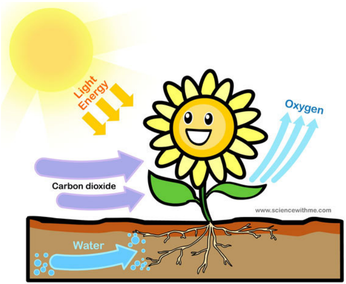
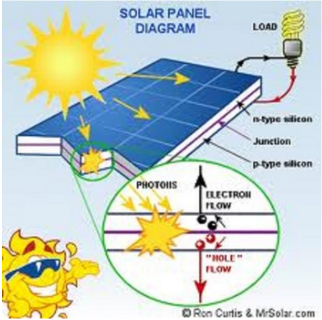
Outline

1. Motivation
2. From DFT to QEDFT
3. Linear response for coupled systems
4. Results

Light matter interaction



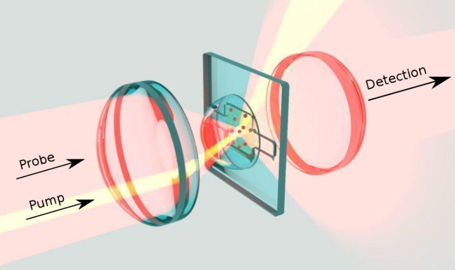
Photosynthesis: A natural process of light and matter interaction[1].



Solarcell: An artificial process of light and matter interaction[2].

[1] www.tes.com
[2] www.examiner.com

Basic concept



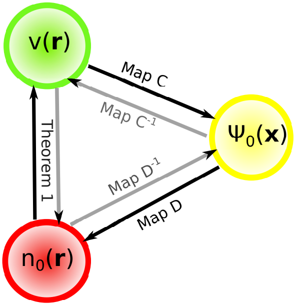
Experimental change: It is now possible to study many-electron systems interacting with the quantized electromagnetic field [3].

- **Well-kown**: The linear response formalism of time-dependent density-functional theory (TDDFT) for fermionic systems only
- **Goal**: An extension of the linear response formalism to coupled electron-photon systems

[3] Nature Photonics, Andreas Masers et. all, *Few-photon coherent nonlinear optics with a single molecule*, 2016, Nature Publishing Group 1749-4893

Density functional theory (DFT)

Hohenberg-Kohn Theorem



Theorem: The one-to-one mapping between external potential and the ground-state density

$$\hat{H}\Psi_j(x_1 \ldots x_N) = E_j \Psi_j(x_1 \ldots x_N).$$

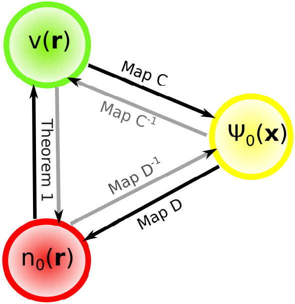
$$\hat{H}_M = -\sum_{j=1}^{N}\left[\frac{\nabla_j^2}{2m} + v(\boldsymbol{r}_j)\right] + \frac{1}{2}\sum_{\substack{j,k \\ j\neq k}}^{N}\frac{e^2}{|\boldsymbol{r}_j - \boldsymbol{r}_k|}$$

$$\Downarrow$$

$$\boxed{v(\boldsymbol{r}) \longleftrightarrow n_0(\boldsymbol{r})}$$

Density functional theory (DFT)

Hohenberg-Kohn Theorem



Theorem: The one-to-one mapping between external potential and the ground-state density

$$\left.\begin{array}{l} \hat{H}\Psi_j(x_1 \dots x_N) = E_j \Psi_j(x_1 \dots x_N). \\ \hat{H}_M = -\sum_{j=1}^{N}\left[\frac{\nabla_j^2}{2m} + v(\boldsymbol{r}_j)\right] + \frac{1}{2}\sum_{\substack{j,k \\ j\neq k}}^{N} \frac{e^2}{|\boldsymbol{r}_j - \boldsymbol{r}_k|} \end{array}\right\}$$

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Density functional theory (DFT)

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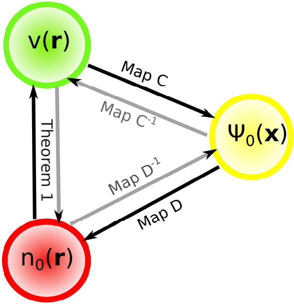
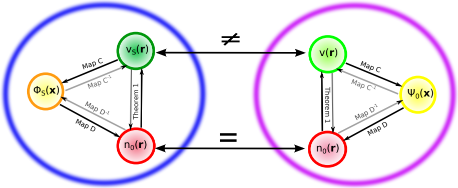


Theorem: The one-to-one mapping between external potential and the ground-state density

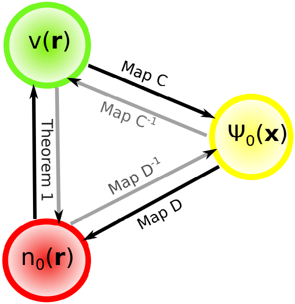
Kohn-Sham system: $W = 0$ Interacting system: $W \neq 0$



Theorem: The ground-state density of an interacting system can be found by solving an effective system of N non-interacting electrons

Extension to quantum-electrodynamical density-functional theory (QEDFT)

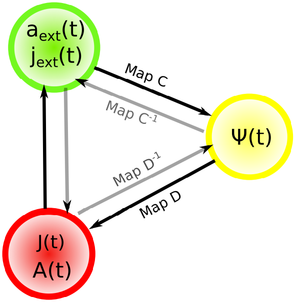
Hohenberg-Kohn theorem



How does the mapping change for QEDFT?

Extension to quantum-electrodynamical density-functional theory (QEDFT)

Bijective mapping



Theorem: The bijective mapping of the external field a_{ext} and current j_{ext} to the internal current J and conjugated field A

$$i\hbar \frac{\partial \Psi(t)}{\partial t} = \hat{H}(t)\Psi(t)$$

$$\hat{H}(t) = \hat{H}_M + \hat{H}_{EM} - \frac{\lambda}{c}\hat{J}\hat{A} - \frac{1}{c}[\hat{J}a_{\text{ext}}(t) + \hat{A}j_{\text{ext}}(t)]$$

$$\hat{H}_{EM} = \sum_j \hbar\omega(\hat{a}_j^{+}\hat{a}_j + \tfrac{1}{2})$$

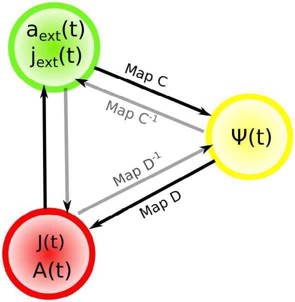
Minimal coupling: $\boldsymbol{p} \rightarrow \boldsymbol{p} - \frac{e}{c}\boldsymbol{A}(\boldsymbol{r}, t)$

$$\Downarrow$$

$$\boxed{\Psi_0 : (a_{ext}, j_{ext}) \leftrightarrow (J, A)}$$

Extension to quantum-electrodynamical density-functional theory (QEDFT)

Bijective mapping



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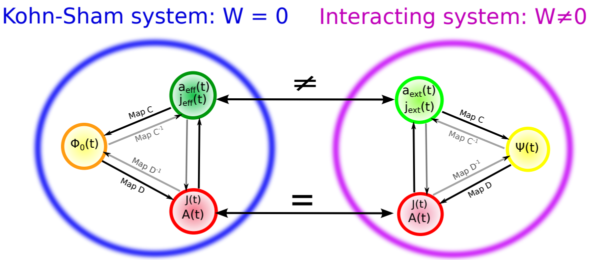
Extension to quantum-electrodynamical density-functional theory (QEDFT)



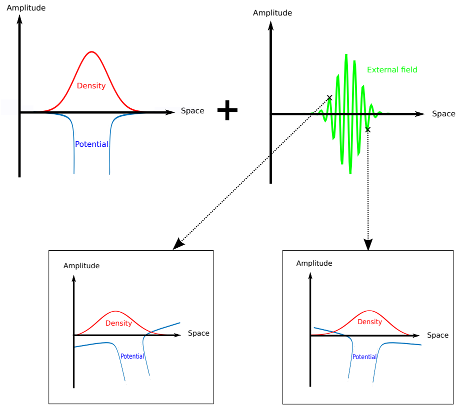
Theorem: The initial pair $J(t), A(t)$ can be found by solving the effective single particle system.[a] [b]

[a] PNAS, Johannes Flick et. all, *Kohn-Sham approach to quantum electrodynamical density-functional theory: Exact time-dependent effective potentials in real space*, vol.112 no.50,2015

[b] Phys.Rev A, Michael Ruggenthaler et. all, *Quantum-electrodynamical density-functional theory: Bridging quantum optics and electronic-structure theory*,vol.90,p.012508,2014

Linear-response formalism

- A reaction of a system to an external perturbation
- Our case: Density change of electrons and photons as a reaction to a weak perturbation $\delta v_{e/pt}(r, t)$
- Can be described to first order $\rightarrow$ linear response functions



Extension of linear-response formalism to coupled systems

- Definition of the linear-response function for the electronic, photonic and coupled case:
  $\chi_{R_e}(rt, r't') := -i\Theta(t-t') \langle\Psi_0| [\hat{n}_{e_H}(rt), \hat{n}_{e_H}(r't')] |\Psi_0\rangle$
  $\chi_{R_{pt}}(rt, r't') := -i\Theta(t-t') \langle\Psi_0| [\hat{n}_{pt_H}(rt), \hat{n}_{pt_H}(r't')] |\Psi_0\rangle$
  $\chi_{R_{pt,e}}(rt, r't') := -i\Theta(t-t') \langle\Psi_0| [\hat{n}_{pt_H}(rt), \hat{n}_{e_H}(r't')] |\Psi_0\rangle$
  $\chi_{R_{e,pt}}(rt, r't') := -i\Theta(t-t') \langle\Psi_0| [\hat{n}_{e_H}(rt), \hat{n}_{pt_H}(r't')] |\Psi_0\rangle$
  with $\hat{n}_e(rt) = \hat{c}^+(rt) \cdot \hat{c}(rt), \quad \hat{n}_{pt}(rt) = \hat{a}^+(rt) + \hat{a}(rt)$
- For application we use
  $\begin{pmatrix} \delta n_e(r,t) \\ \delta n_{pt}(r,t) \end{pmatrix} =$
  $\int dt' \int dr' \begin{pmatrix} \chi_{e,e}(rt, r't') & \chi_{e,pt}(rt, r't') \\ \chi_{pt,e}(rt, r't') & \chi_{pt,pt}(rt, r't') \end{pmatrix} \cdot \begin{pmatrix} \delta v_e(r', t') \\ \delta v_{pt}(r', t') \end{pmatrix}$

Lehmann-representation for coupled systems

- For calculations we use eigenenergy-representation of the response function
- Eigenvalue problem $H\,|\Psi_j\rangle = E_j\,|\Psi_j\rangle$, with H being a coupled electron-photon Hamiltonian
- We rewrite the linear response functions in space and time according to

$$\chi_{R_e}(r,r',t-t') = -\Theta(t-t')\sum_S e^{-i\Omega_{N,S}(t-t')} f_{e_{N,S}}(r)\cdot f^*_{e_{N,S}}(r') + c.c$$

$$\chi_{R_{pt}}(r,r',t-t') = -\Theta(t-t')\sum_S e^{-i\Omega_{N,S}(t-t')} f_{pt_{N,S}}(r)\cdot f^*_{pt_{N,S}}(r') + c.c$$

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Linear response of the Kohn-Sham system

TDDFT

- Connection: linear-response formalism and TDDFT $\rightarrow$ linear response for the Kohn-Sham system
- A relation between interacting and non-interacting linear response:

$$f_{Hxc}(\boldsymbol{r},\boldsymbol{r}',\omega) = \chi_s^{-1}(\boldsymbol{r},\boldsymbol{r}',\omega) - \chi^{-1}(\boldsymbol{r},\boldsymbol{r}',\omega)$$

QEDFT

- Response function now takes a matix form:

$$\chi(\boldsymbol{r}t,\boldsymbol{r}'t') = \begin{pmatrix} \chi_{\mathrm{e}}(\boldsymbol{r}t,\boldsymbol{r}'t') & \chi_{\mathrm{e,pt}}(\boldsymbol{r}t,\boldsymbol{r}'t') \\ \chi_{\mathrm{pt,e}}(\boldsymbol{r}t,\boldsymbol{r}'t') & \chi_{\mathrm{pt}}(\boldsymbol{r}t,\boldsymbol{r}'t') \end{pmatrix}$$

$$\chi_s(\boldsymbol{r},t,\boldsymbol{x},\tau) = \begin{pmatrix} \chi_{s_{\mathrm{e}}}(\boldsymbol{r},t,\boldsymbol{x},\tau) & 0 \\ 0 & \chi_{s_{\mathrm{pt}}}(\boldsymbol{r},t,\boldsymbol{x},\tau) \end{pmatrix}$$

$$f_{Hxc}(\boldsymbol{x}\tau,\boldsymbol{x}'\tau') = \begin{pmatrix} f_{Hxc_{\mathrm{e}}}(\boldsymbol{x}\tau,\boldsymbol{x}'\tau') & f_{Hxc_{\mathrm{e,pt}}}(\boldsymbol{x}\tau,\boldsymbol{x}'\tau') \\ f_{Hxc_{\mathrm{pt,e}}}(\boldsymbol{x}\tau,\boldsymbol{x}'\tau') & f_{Hxc_{\mathrm{pt}}}(\boldsymbol{x}\tau,\boldsymbol{x}'\tau') \end{pmatrix}$$

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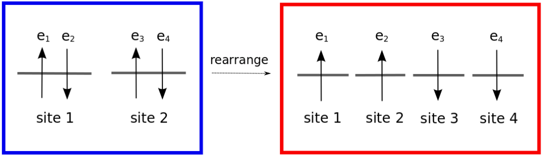
Lattice model

Electronic model:



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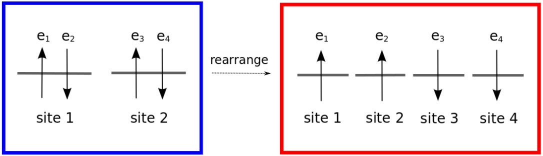
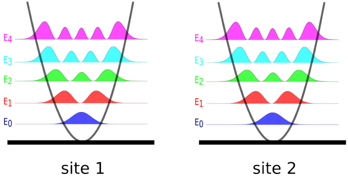


Photonic model:



Hamiltonian

Hamiltonian for two sites and two Fock-number states:

$$H = H_e + H_{pt} + H_{e,pt} + H_{ext} =$$

$$-t_0(c^+_{1\uparrow}c_{2\uparrow} + c^+_{1\downarrow}c_{2\downarrow} + c^+_{2\uparrow}c_{1\uparrow} + c^+_{2\downarrow}c_{1\downarrow})+$$

$$2t_0(c^+_{1\uparrow}c_{1\uparrow} + c^+_{1\downarrow}c_{1\downarrow} + c^+_{2\uparrow}c_{2\uparrow} + c^+_{2\downarrow}c_{2\downarrow})+$$

$$\hbar\omega_{pt}(a_1^+ a_1 + a_2^+ a_2 + 1)+$$

$$\lambda[(c^+_{1\uparrow}c_{1\uparrow} + c^+_{1\downarrow}c_{1\downarrow})(a_1^+ + a_1)+(c^+_{2\uparrow}c_{2\uparrow} + c^+_{2\downarrow}c_{2\downarrow})(a_2^+ + a_2)]+$$

$$\left(\hat{x}_{1\uparrow}(c^+_{1\uparrow}c_{1\uparrow}) + \hat{x}_{1\downarrow}(c^+_{1\downarrow}c_{1\downarrow}) + \hat{x}_{2\uparrow}(c^+_{2\uparrow}c_{2\uparrow}) + \hat{x}_{2\downarrow}(c^+_{2\downarrow}c_{2\downarrow})\right)\left(E_0 e^{\frac{-(t-t_0)^2}{\sigma^2}} \sin(\omega t)\right)$$

$$+\left(\hat{x}_1(a_1^+ + a_1) + \hat{x}_2(a_2^+ + a_2)\right)\left(J_0 e^{\frac{-(t-t_0)^2}{\sigma^2}} \sin(\omega t)\right)$$

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\hbar\omega_{pt}(a^+_1 a_1 + a^+_2 a_2 + 1) + \\
\lambda[(c^+_{1\uparrow}c_{1\uparrow} + c^+_{1\downarrow}c_{1\downarrow})(a^+_1 + a_1) + (c^+_{2\uparrow}c_{2\uparrow} + c^+_{2\downarrow}c_{2\downarrow})(a^+_2 + a_2)] + \\
\left(\hat{x}_{1\uparrow}(c^+_{1\uparrow}c_{1\uparrow}) + \hat{x}_{1\downarrow}(c^+_{1\downarrow}c_{1\downarrow}) + \hat{x}_{2\uparrow}(c^+_{2\uparrow}c_{2\uparrow}) + \hat{x}_{2\downarrow}(c^+_{2\downarrow}c_{2\downarrow})\right)\left(E_0 e^{\frac{-(t-t_0)^2}{\sigma^2}} \sin(\omega t)\right) \\
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2t_0(c^+_{1\uparrow}c_{1\uparrow} + c^+_{1\downarrow}c_{1\downarrow} + c^+_{2\uparrow}c_{2\uparrow} + c^+_{2\downarrow}c_{2\downarrow}) + \\
\hbar\omega_{pt}(a^+_1 a_1 + a^+_2 a_2 + 1) + \\
\lambda[(c^+_{1\uparrow}c_{1\uparrow} + c^+_{1\downarrow}c_{1\downarrow})(a^+_1 + a_1) + (c^+_{2\uparrow}c_{2\uparrow} + c^+_{2\downarrow}c_{2\downarrow})(a^+_2 + a_2)] + \\
\left(\hat{x}_{1\uparrow}(c^+_{1\uparrow}c_{1\uparrow}) + \hat{x}_{1\downarrow}(c^+_{1\downarrow}c_{1\downarrow}) + \hat{x}_{2\uparrow}(c^+_{2\uparrow}c_{2\uparrow}) + \hat{x}_{2\downarrow}(c^+_{2\downarrow}c_{2\downarrow})\right)\left(E_0 e^{\frac{-(t-t_0)^2}{\sigma^2}} \sin(\omega t)\right) \\
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\hbar\omega_{pt}(a^+_1 a_1 + a^+_2 a_2 + 1) + \\
\lambda[(c^+_{1\uparrow}c_{1\uparrow} + c^+_{1\downarrow}c_{1\downarrow})(a^+_1 + a_1) + (c^+_{2\uparrow}c_{2\uparrow} + c^+_{2\downarrow}c_{2\downarrow})(a^+_2 + a_2)] + \\
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$$2t_0(c^+_{1\uparrow}c_{1\uparrow} + c^+_{1\downarrow}c_{1\downarrow} + c^+_{2\uparrow}c_{2\uparrow} + c^+_{2\downarrow}c_{2\downarrow})+$$
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$$\lambda[(c^+_{1\uparrow}c_{1\uparrow} + c^+_{1\downarrow}c_{1\downarrow})(a^+_1 + a_1)+(c^+_{2\uparrow}c_{2\uparrow} + c^+_{2\downarrow}c_{2\downarrow})(a^+_2 + a_2)]+$$
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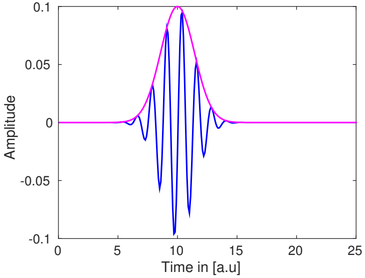
$$+\left(\hat{x}_1(a^+_1 + a_1) + \hat{x}_2(a^+_2 + a_2)\right)\left(J_0 e^{\frac{-(t-t_0)^2}{\sigma^2}}\sin(\omega t)\right)$$

External perturbation

$$H_{ext} =$$
$$\left(\hat{x}_{1\uparrow}(c^{+}_{1\uparrow}c_{1\uparrow}) + \hat{x}_{1\downarrow}(c^{+}_{1\downarrow}c_{1\downarrow}) + \hat{x}_{2\uparrow}(c^{+}_{2\uparrow}c_{2\uparrow}) + \hat{x}_{2\downarrow}(c^{+}_{2\downarrow}c_{2\downarrow})\right)\left(E_0 e^{\frac{-(t-t_0)^2}{\sigma^2}}\sin(\omega t)\right)$$
$$+\left(\hat{x}_1(a^{+}_1 + a_1) + \hat{x}_2(a^{+}_2 + a_2)\right)\left(J_0 e^{\frac{-(t-t_0)^2}{\sigma^2}}\sin(\omega t)\right)$$



Overview

H_e	H_{pt}	H	v_{ext}	j_{ext}	linear response analytic	linear response numeric	time-propagation	notation	case
x			x		x	x	x	consistency check	[1]
	x			x	x	x	x	consistency check	[2]
		x	x			x	x	comparison with case 1	[3]
		x		x		x	x	comparison with case 2	[4]
		x	x	x		x		Pump-probe pulse	[5]

Overview

H_e	H_{pt}	H	v_{ext}	j_{ext}	linear response analytic	linear response numeric	time-propagation	notation	case
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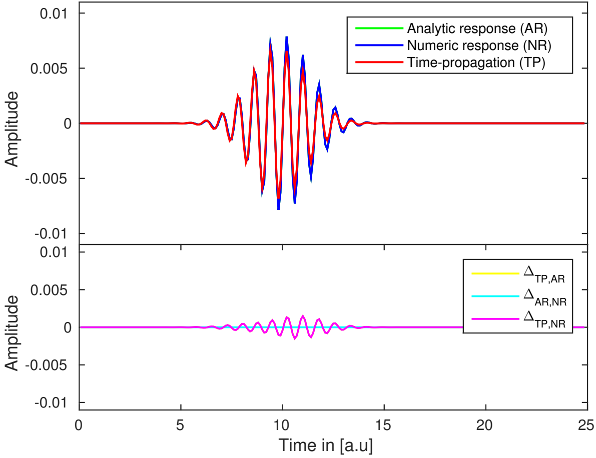
	x			x	x	x	x	consistency check	[2]
		x	x			x	x	comparison with case 1	[3]
		x		x		x	x	comparison with case 2	[4]
		x	x	x		x		Pump-probe pulse	[5]

Consistency check for the electronic system (case[1])

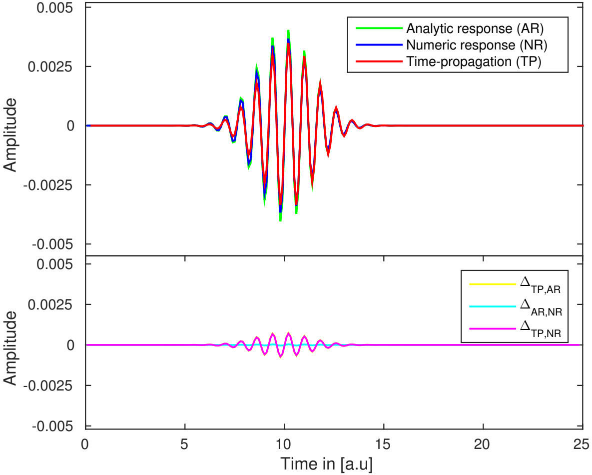


Paramerters in [a.u]: $\omega = 8, \quad \sigma = 2, \quad E_0 = 0.1$

Overview

H_e	H_{pt}	H	v_{ext}	j_{ext}	linear response analytic	linear response numeric	time-propagation	notation	case
x			x		x	x	x	consistency check	[1]
	x			x	x	x	x	consistency check	[2]
		x	x			x	x	comparison with case 1	[3]
		x		x		x	x	comparison with case 2	[4]
		x	x	x		x		Pump-probe pulse	[5]

Consistency check for the photonic system (case[2])



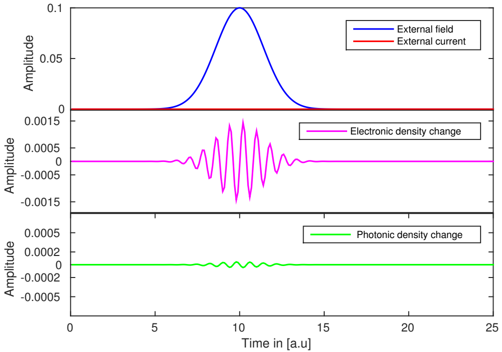
Paramerters in [a.u]: $\omega = 8, \quad \sigma = 2, \quad E_0 = 0.1, \quad \omega_{pt} = 4$

Overview

H_e	H_{pt}	H	v_{ext}	j_{ext}	linear response analytic	linear response numeric	time-propagation	notation	case
x			x		x	x	x	consistency check	[1]
	x			x	x	x	x	consistency check	[2]
		x	x			x	x	comparison with case 1	[3]
		x		x		x	x	comparison with case 2	[4]
		x	x	x		x		Pump-probe pulse	[5]

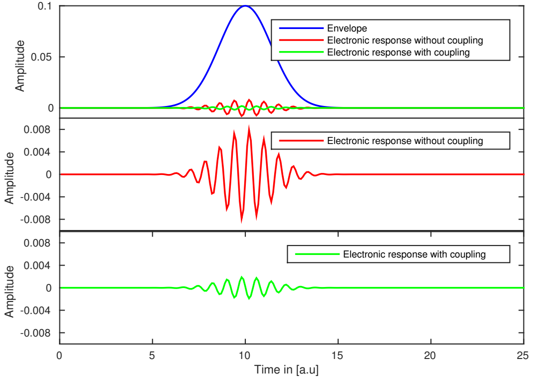
Electronic linear response of the electron- photon coupled system (case[3])

Response of the coupled system with $\delta v_{ext} \neq 0$ and $\delta j_{ext} = 0$:



Although there is **no explicit external perturbation for the photons** themselves (red), a **photonic linear response** (green) is generated!

Comparison of electronic linear response of the coupled and uncoupled system



Amplitude difference

$\Downarrow$

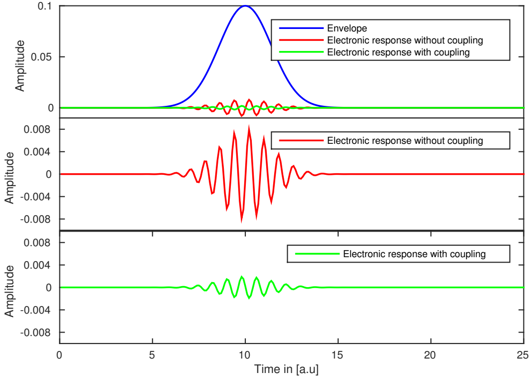
Energy conservation

Phase shift

$\Downarrow$

Time-delay of displacement of charged particles and the radiation of an electro-magnetic signal

Comparison of electronic linear response of the coupled and uncoupled system



Amplitude difference

$\Downarrow$

Energy conservation

Phase shift

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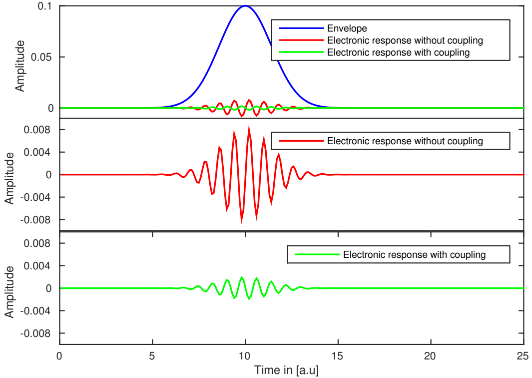
Time-delay of displacement of charged particles and the radiation of an electro-magnetic signal

Comparison of electronic linear response of the coupled and uncoupled system



Amplitude difference

$\Downarrow$

Energy conservation

Phase shift

$\Downarrow$

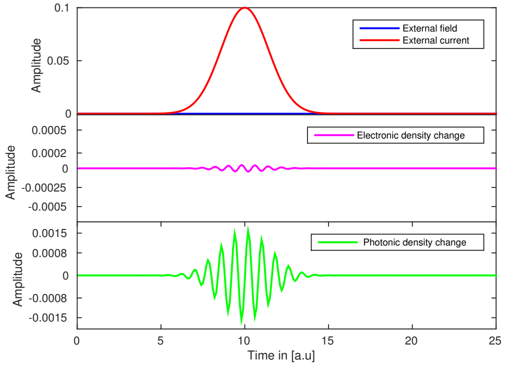
Time-delay of displacement of charged particles and the radiation of an electro-magnetic signal

Overview

H_e	H_{pt}	H	v_{ext}	j_{ext}	linear response analytic	linear response numeric	time-propagation	notation	case
x			x		x	x	x	consistency check	[1]
	x			x	x	x	x	consistency check	[2]
		x	x			x	x	comparison with case 1	[3]
		x		x		x	x	comparison with case 2	[4]
		x	x	x		x		Pump-probe pulse	[5]

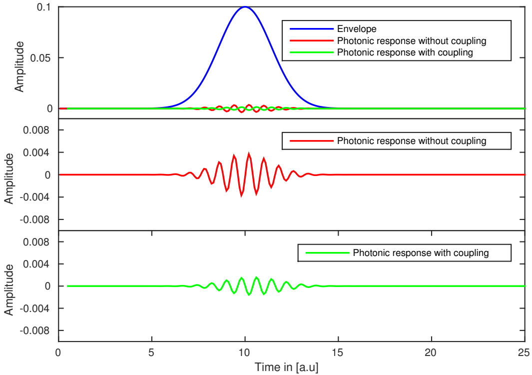
Photonic linear response of the electron- photon coupled system (case[4])

Response of the coupled system with $\delta v_{ext} = 0$ and $\delta j_{ext} \neq 0$:



Although there is **no explicit external perturbation for the electrons** themselves (blue), an **electronic linear response** (purple) is generated!

Comparison of photonic linear response of the coupled and uncoupled system



Amplitude difference

$\Downarrow$

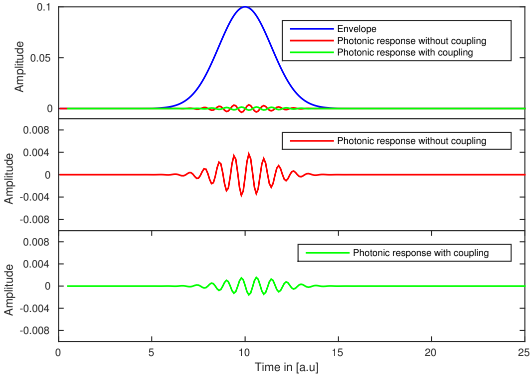
Energy conservation

Phase shift

$\Downarrow$

Time-delay of displacement of charged particles and the radiation of an electro-magnetic signal

Comparison of photonic linear response of the coupled and uncoupled system



Amplitude difference

$\Downarrow$

Energy conservation

Phase shift

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Time-delay of displacement of charged particles and the radiation of an electro-magnetic signal

Overview

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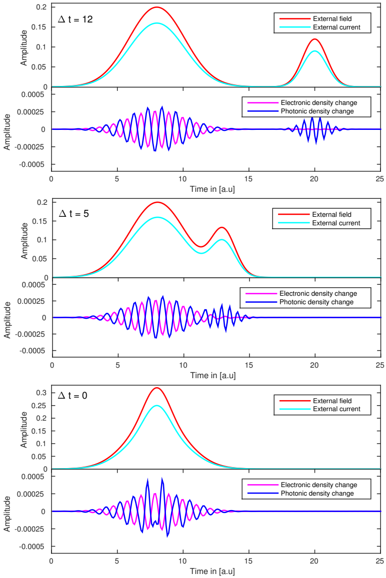
x			x		x	x	x	consistency check	[1]
	x			x	x	x	x	consistency check	[2]
		x	x			x	x	comparison with case 1	[3]
		x		x		x	x	comparison with case 2	[4]
		x	x	x		x		Pump-probe pulse	[5]

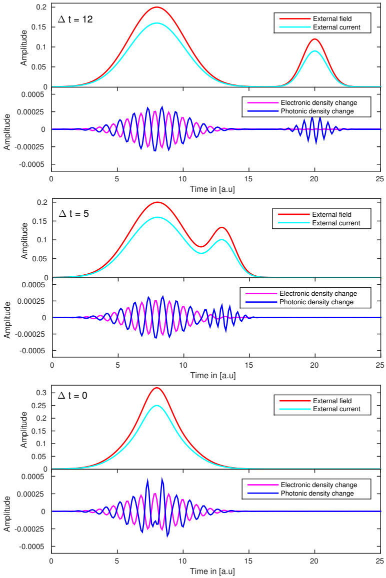
Pump-probe experiment (case[5])



Pump-Probe Experiment:

- Two laser pulses applied with time-delay
- Each pulse contains field and current
- For calculations the electron-photon coupled Hamiltonian is used
- **Parameters:**
  Coupling strength $\lambda = 0.6$
  Hopping parameter $t_0 = 1$
  Photonic frequency $\omega_{pt} = 4$

Pump-probe experiment (case[5])



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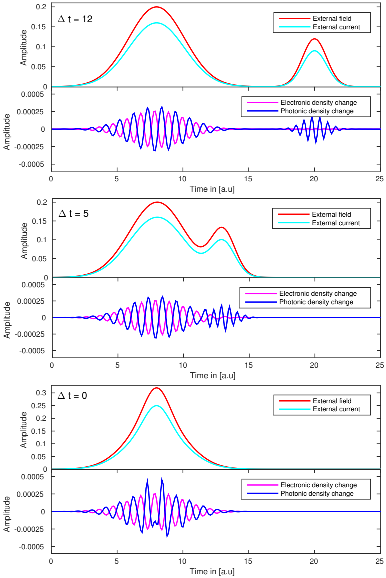
Pump-probe experiment (case[5])



Pump-Probe Experiment:

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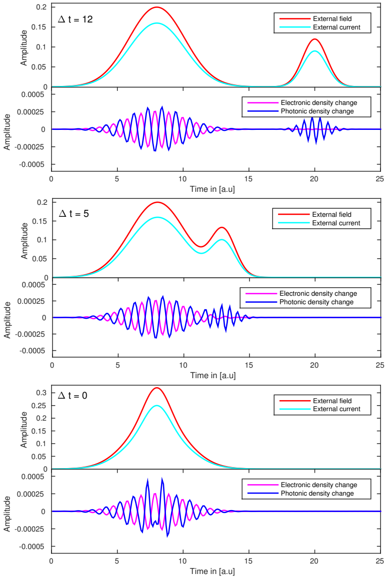
Pump-probe experiment (case[5])



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- **Concluded steps:**
  1. Extension of the linear response formalism to coupled system
  2. Successful consistency check
  3. Investigate different coupling cases
  4. First approaches to experimental scenarion (Pump-probe experiment)
- **Next steps:**
  1. Extensions for more sites
  2. Introducing formalism for different particle numbers
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