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Published in:
Proceedings of the Conference Progress in Nonequilibrium Green's Functions II

NONEQUILIBRIUM GREEN FUNCTIONS IN TIME-DEPENDENT CURRENT-DENSITY-FUNCTIONAL THEORY*

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We give an overview of the underlying concepts of time-dependent current-density-functional theory (TDCDFT). We show how the basic equations of TDCDFT can be elegantly derived using the time contour method of nonequilibrium Green function theory. We further demonstrate how the formalism can be used to derive explicit equations for the exchange-correlation vector potentials and integral kernels for the Kohn-Sham equations and their linearized form.

1. Introduction

Time-dependent density-functional theory (TDDFT) is a method for calculating properties of many-electron systems in time-dependent external fields (for reviews see [1,2]). In TDDFT one introduces a noninteracting system with the same density $n(\mathbf{r}t)$ as the true system. This system is called the Kohn-Sham system and its external potential, which is a functional of the density, is denoted as $v_s(\mathbf{r}t)$. Once an approximate functional is given the Kohn-Sham equations can be solved self-consistently as has been done very successfully. Nevertheless, the theory has the limitation that it can not deal with transverse fields. This limitation is lifted in time-dependent current-density-functional theory (TDCDFT), [3] whose basic variable is the current-density in addition to the density and in which an effective vector potential $\mathbf{A}_s$ appears as well as a scalar potential. This theory has shown to be very successful in describing macroscopic polarization in solids [4], plasmon line widths [5] and polarizabilities of polymers [6]. In this work we

*based on a poster presented at the conference "progress in nonequilibrium greens functions, dresden, germany, 19.-22. august 2002"

will describe how systematic approximations for $\mathbf{A}_s$ can be obtained from perturbation theory on the basis of Keldysh Green functions.

2. The field-matter Hamiltonian

The full Hamiltonian in the Heisenberg picture for a quantized electromagnetic field and nonrelativistic electrons in the presence of a classical external electromagnetic field $(\phi^{\mathbf{ext}}, \mathbf{A}^{\mathbf{ext}})$ is given by [8,7]

$$\hat{H} = \frac{1}{8\pi}\int d^3r((\nabla\hat{\phi} + \partial_t\hat{\mathbf{A}})^2 + c^2(\nabla\times\hat{\mathbf{A}})^2) + \sum_\sigma \int d^3r\left(\frac{1}{2}\hat{\psi}^\dagger_\sigma(-i\nabla - e(\hat{\mathbf{A}} + \mathbf{A}^{\rm ext}))^2\hat{\psi}_\sigma + e\hat{\psi}^\dagger_\sigma\hat{\psi}_\sigma\phi^{\rm ext}\right) \quad (1)$$

where σ is a spin index, e is the electron charge and we use the Coulomb gauge $\nabla\cdot\hat{\mathbf{A}} = \nabla\cdot\mathbf{A}^{\rm ext} = 0$. We formally obtain the operator equations

$$\hat{\phi}(\mathbf{r}t) = e\int d^3r'\frac{\hat{n}(\mathbf{r}'t)}{|\mathbf{r}-\mathbf{r}'|} \quad (2)$$

$$\hat{\mathbf{A}}(\mathbf{r}t) = \frac{e}{c^2}\int d^3r'\frac{\hat{\mathbf{j}}^T(\mathbf{r}', t-|\mathbf{r}-\mathbf{r}'|/c)}{|\mathbf{r}-\mathbf{r}'|} \quad (3)$$

where $\hat{n}$ is the number density operator and $\mathbf{j}^T$ is the transverse part of the particle current operator $\hat{\mathbf{j}}(\mathbf{r}t) = \hat{\mathbf{j}}_p(\mathbf{r}t) - e\hat{n}(\mathbf{r}t)\hat{\mathbf{A}}^{\rm tot}(\mathbf{r}t)$ where we defined the total vector potential by $\hat{\mathbf{A}}^{\rm tot} = \hat{\mathbf{A}} + \mathbf{A}^{\rm ext}$ and the density and paramagnetic current (Heisenberg) operators by

$$\hat{n}(\mathbf{r}t) = \sum_\sigma \hat{\psi}^\dagger_\sigma(\mathbf{r}t)\hat{\psi}_\sigma(\mathbf{r}t) \quad (4)$$

$$\hat{\mathbf{j}}_p(\mathbf{r}t) = \sum_\sigma \frac{1}{2i}(\hat{\psi}^\dagger_\sigma(\mathbf{r}t)\nabla\hat{\psi}_\sigma(\mathbf{r}t) - [\nabla\hat{\psi}^\dagger_\sigma(\mathbf{r}t)]\hat{\psi}_\sigma(\mathbf{r}t)) \quad (5)$$

If we now insert the explicit form of operator $\hat{\phi}$ into the full Hamiltonian and replace $\hat{\mathbf{A}}$ in the mean field approximation with its expectation value $\langle\hat{\mathbf{A}}\rangle$ and treat this field classically, we obtain (using Gauss' theorem to transform $(\nabla\hat{\phi})^2$ to $\hat{\phi}\nabla^2\hat{\phi}$) the following Hamiltonian in the Schrödinger picture for the electrons

$$\hat{H} = \sum_\sigma \frac{1}{2}\int d^3r\hat{\psi}^\dagger_\sigma(\mathbf{r})(-i\nabla - e\langle\hat{\mathbf{A}}^{\rm tot}(\mathbf{r}t)\rangle)^2\hat{\psi}_\sigma(\mathbf{r}) + e\hat{n}\phi^{\rm ext}(\mathbf{r}t) + \sum_{\sigma,\sigma'}\frac{e^2}{2}\int d^3rd^3r'\frac{\hat{\psi}^\dagger_\sigma(\mathbf{r})\hat{\psi}^\dagger_{\sigma'}(\mathbf{r}')\hat{\psi}_{\sigma'}(\mathbf{r}')\hat{\psi}_\sigma(\mathbf{r})}{|\mathbf{r}-\mathbf{r}'|} \quad (6)$$

This is the Hamiltonian that will be used as the basis of our theory. In the following we put $e = -1$ and we will neglect the induced field $\langle\hat{\mathbf{A}}\rangle$ such

that $\langle \hat{\mathbf{A}}^{\rm tot} \rangle = \hat{\mathbf{A}}^{\rm ext}$ and call this field $\mathbf{A}(\mathbf{r}t)$. Note that this is allowed in finite systems, but not in infinite solids where there will be a macroscopic contribution to the induced field [4]. We will also choose a gauge in which $\phi^{\rm ext}$ is the potential of the nuclear framework of a solid or molecule. All other potentials (externally applied or effective) will be described by a vector potential $\mathbf{A}$ which in general will also contain longitudinal components.

3. The fundamental equations of TDCDFT

We start out by defining the Schwinger-Keldysh contour [9,10,11,12] . This is a parametrization of real time $t(\tau)$ by an underlying pseudotime parameter such that if τ runs from a certain initial τ_i to a final τ_f then the real time runs from t_0 to t_1 and back to t_0 (in practice we will often choose $t_0 = -\infty$ and $t_1 = +\infty$). We then define a generating functional, resembling the partition function of statistical mechanics:

$$\tilde{F}[\mathbf{A}] = i \ln \langle \Psi_0 | V(\tau_f, \tau_i) | \Psi_0 \rangle \tag{7}$$

where V is the τ- or contour ordered evolution operator of the system

$$V(\tau_2, \tau_1) = T_C \exp\left[-i \int_{\tau_1}^{\tau_2} d\tau t'(\tau) \hat{H}(\tau)\right] \tag{8}$$

with $t'(\tau) = dt/d\tau$ and where T_C denotes ordering in τ, $\hat{H}$ is the Hamiltonian of Eq.(6) and the state $|\Psi_0\rangle$ is the initial state. A point on the contour will be denoted as $\bar{1} = \mathbf{r}_1 \tau_1$, whereas a point in physical time is denoted as $1 = \mathbf{r}_1 t_1$. Here we extend the original definition of the Hamiltonian and allow $\mathbf{A}(\bar{1})$ to take different values of the forward and backward parts of the contour. Functional derivatives will eventually be evaluated at physical vector potentials with identical values on both sides of the contour. To evaluate the functional derivative we use

$$\delta V(\tau_2, \tau_1)/\delta \mathbf{A}(\bar{3}) = -iV(\tau_2, \tau)\hat{\mathbf{j}}(\bar{3})V(\tau, \tau_1) \tag{9}$$

where $\tau_1 < \tau < \tau_2$. These equations follow directly from the Schrödinger equation. Using these equations we find

$$\delta \tilde{F}/\delta \mathbf{A}(\bar{1}) = \langle \hat{\mathbf{j}}_H(\bar{1}) \rangle = \mathbf{j}(\bar{1}) \tag{10}$$

where we defined the Heisenberg representation of an operator as usual by $\hat{O}_H(\tau) = V(\tau_i, \tau)\hat{O}V(\tau, \tau_i)$ and the expectation value by

$$\langle \hat{O}_H(\tau) \rangle = \frac{\langle \Psi_0 | T_C[V(\tau_f, \tau_i)\hat{O}_H(\tau)] | \Psi_0 \rangle}{\langle \Psi_0 | V(\tau_f, \tau_i) | \Psi_0 \rangle} \tag{11}$$

Therefore the derivative of $\tilde{F}$ at the physical potential $\mathbf{A}$ is the gauge invariant current $\mathbf{j}(\mathbf{r}t)$ of the system in the external field $\mathbf{A}(\mathbf{r}t)$. We can now calculate higher order response functions by repeated differentiation. To not overcrowd notation we drop the subindex H from the operators. The current-current response function $\chi_{\mu\nu}$ is then given by

$$\chi_{\mu\nu}(\bar{1},\bar{2}) = \frac{\delta^2 \tilde{F}}{\delta A_\mu(\bar{1})\delta A_\nu(\bar{2})}$$

$$= \delta_{\mu\nu} n(\bar{1})\delta_C(t_1 - t_2) - i\langle T_C[\Delta\hat{\mathbf{j}}_{p,\mu}(\bar{1})\Delta\hat{\mathbf{j}}_{p,\nu}(\bar{2})]\rangle \tag{12}$$

where $\delta_C(t_1 - t_2) = \delta(\tau_1 - \tau_2)/t'(\tau_1)$ is the contour delta function [11] and where the current-density fluctuation operator $\Delta\hat{\mathbf{j}}_{p,\mu}(\bar{1}) = \hat{\mathbf{j}}_{p,\mu}(\bar{1}) - \langle\hat{\mathbf{j}}_{p,\mu}(\bar{1})\rangle$ enters due to the derivatives of the denominator in Eq.(10). The response function is a symmetric function of its arguments as it should (being a second order functional derivative) and can be regarded as a integral kernel in pseudotime. It will however become a retarded function acting in physical time as can be seen from the current response $\delta\mathbf{j}(\mathbf{r}t)$ due to a physical potential variation $\delta\mathbf{A}(\mathbf{r}t)$:

$$\delta j_\mu(1) = \sum_\nu \int_C d\bar{2}\chi_{\mu\nu}(\bar{1},\bar{2})\delta A_\nu(2) = \sum_\nu \int d2\chi_{R,\mu\nu}(1,2)\delta A_\nu(2) \tag{13}$$

Here the function $\chi_{R,\mu\nu}$ is the usual retarded response function

$$\chi_{R,\mu\nu}(1,2) = n_0(\mathbf{r}_1)\delta_{\mu\nu}\delta(t_1 - t_2)\delta(\mathbf{r}_1 - \mathbf{r}_2)$$
$$- i\theta(t_1 - t_2)\langle\Psi_0|[\hat{\mathbf{j}}_{p,\mu}(1),\hat{\mathbf{j}}_{p,\nu}(2)]|\Psi_0\rangle \tag{14}$$

as it usually appears in response theory. The outlined procedure applies to all higher order derivatives as well, i.e. all higher order response functions are symmetric functions in pseudotime and become causal or retarded functions in physical time. This feature has been used to resolve a recent paradox in TDDFT [13].

We now want to use the current $\mathbf{j}(\mathbf{r}\tau)$ as our basic variable and we perform a Legendre transform and define

$$F[\mathbf{j}] = -\tilde{F}[\mathbf{A}] + \int_C d\bar{1}\mathbf{j}(\bar{1})\cdot\mathbf{A}(\bar{1}) \tag{15}$$

and hence $\delta F/\delta\mathbf{j}(\bar{1}) = \mathbf{A}(\bar{1})$. The Legendre transformation assumes that there is a one-to-one relation between $\mathbf{j}(\bar{1})$ and $\mathbf{A}(\bar{1})$ such that Eq. (10) is invertible (up to gauge). One can prove, by a direct generalization of the proof given for the density-density response function in ref.[2], that the Keldysh current-current reponse function is invertible for switch-on processes. We

now define an action functional for a noninteracting system with the Hamiltonian $\hat{H}_s$ obtained from Eq.(6) by removal of the Coulomb interaction and replacement of $\mathbf{A}$ by an effective vector potential $\mathbf{A}_s$ (subindex s refers to single-particle, as the system can now be descibed by single-particle equations).

$$\hat{H}_s = \sum_\sigma \frac{1}{2} \int d^3r \hat{\psi}^\dagger_\sigma(\mathbf{r})(-i\nabla - e\hat{\mathbf{A}}_s(\mathbf{r}t))^2 \hat{\psi}_\sigma(\mathbf{r}) + e\hat{n}\phi^{\rm ext}(\mathbf{r}t) \tag{16}$$

We then define

$$\tilde{F}_s[\mathbf{A}_s] = i \ln\langle\Phi_0|V_s(\tau_f,\tau_i)|\Phi_0\rangle \tag{17}$$

The evolution operator $V_s(\tau_f,\tau_i)$ is defined similarly as in Eq.(8) with $\hat{H}$ replaced by $\hat{H}_s$. The initial wave function Φ_0 at $t = t_0$ is a noninteracting state and will often be a Slater determinant. We can now do again a Legendre transform and define

$$F_s[\mathbf{j}] = -\tilde{F}_s[\mathbf{A}_s] + \int_C d\bar{1}\mathbf{j}(\bar{1}) \cdot \mathbf{A}_s(\bar{1}) \tag{18}$$

The exchange-correlation part F_{xc} of the action functional is then defined by

$$F[\mathbf{j}] = F_s[\mathbf{j}] - F_{xc}[\mathbf{j}] - \frac{1}{2}\int_C d\bar{1} \int_C d\bar{2} v(\bar{1},\bar{2}) n(\bar{1}) n(\bar{2}) \tag{19}$$

where $v(\bar{1},\bar{2}) = \delta_C(t_1 - t_2)/|\mathbf{r}_1 - \mathbf{r}_2|$. The Keldysh density n is a functional of the initial state and the current $\mathbf{j}$ through the continuity equation. Functional differentiation of Eq. (19) with respect to $\mathbf{j}(\bar{1})$ yields

$$\mathbf{A}(\bar{1}) = \mathbf{A}_s(\bar{1}) - \mathbf{A}_{xc}(\bar{1}) - \mathbf{A}_H(\bar{1}) \tag{20}$$

$$\partial_t \mathbf{A}_H(\bar{1}) = -\nabla \int d^3r' \frac{n(\bar{1})}{|\mathbf{r} - \mathbf{r}'|} \tag{21}$$

$$\mathbf{A}_{xc}(\bar{1}) = \delta F_{xc}/\delta\mathbf{j}(\bar{1}) \tag{22}$$

where $\mathbf{A}_H$ and $\mathbf{A}_{xc}$ are the Hartree and exchange-correlation vector potential. By construction the vector potential $\mathbf{A}_s$ of the noninteracting system yields the same density as the vector potential $\mathbf{A}$ in the fully interacting system. The noninteracting system is thus to be identified with the time-dependent Kohn-Sham system. Let us now see how the current-current response function can be obtained from this formalism. We can write

$$\begin{aligned}\frac{\delta j_\mu(\bar{1})}{\delta A_\nu(\bar{2})} &= \sum_\lambda \int_C d3 \frac{\delta j_\mu(\bar{1})}{\delta A_{s,\lambda}(\bar{3})} \frac{\delta A_{s,\lambda}(\bar{3})}{\delta A_\nu(\bar{2})} = \frac{\delta j_\mu(\bar{1})}{\delta A_{s,\nu}(\bar{2})} \\ &+ \sum_{\lambda\kappa} \int_C d3d4 \frac{\delta j_\mu(\bar{1})}{\delta A_{s,\lambda}(\bar{3})} \frac{\delta(A_{H,\lambda}(\bar{3}) + A_{xc,\lambda}(\bar{3}))}{\delta j_\kappa(\bar{4})} \frac{\delta j_\kappa(\bar{4})}{\delta A_\nu(\bar{2})}\end{aligned} \tag{23}$$

Hence we obtain

$$\chi_{\mu\nu}(\bar{1},\bar{2}) = \chi_{s,\mu\nu}(\bar{1},\bar{2}) + \sum_{\lambda\kappa}\int_C d3d4\chi_{s,\mu\lambda}(\bar{1},\bar{3})(f_{H,\lambda\kappa}(\bar{3},\bar{4}) + f_{xc,\lambda\kappa}(\bar{3},\bar{4}))\chi_{\kappa\nu}(\bar{4},\bar{2}) \quad (24)$$

where $\chi_{s,\mu\nu}$ is the current-current response function of the Kohn-Sham system and where $f_{H,\mu\nu}$ and $f_{xc,\mu\nu}$ are the functional derivatives of $\mathbf{A}_H$ and $\mathbf{A}_{xc}$ with respect to $\mathbf{j}$. We have therefore found a relation between the full and the Kohn-Sham current-current response function. An explicit form of $f_{H,\mu\nu}$ can be obtained from Eq.(21). Eq.(24) is the basic equation of time-dependent current response theory. When transformed to physical time this equations retains the same form with all response functions and kernels replaced by their retarded equivalent.

4. Perturbation theory

Let us now see how we can obtain $\mathbf{A}_{xc}$ and $f_{xc,\mu\nu}$ from perturbation theory. The potential $\mathbf{A}_{xc}$ is determined by the requirement that the Kohn-Sham and real system have the same current, i.e.

$$\langle\hat{\mathbf{j}}_p(\mathbf{r}_1)\rangle + \langle\hat{n}(\bar{1})\rangle\mathbf{A}(\bar{1}) = \langle\hat{\mathbf{j}}_p(\mathbf{r}_1)\rangle_0 + \langle\hat{n}(\bar{1})\rangle_0\mathbf{A}_s(\bar{1}) \quad (25)$$

where the subindex 0 denotes the expectation value with respect to the Kohn-Sham state $|\Phi_0\rangle$. Note that by construction $\langle\hat{n}\rangle_0 = \langle\hat{n}\rangle = n$. To connect the fully interacting initial state $|\Psi_0\rangle$ to the Kohn-Sham state $|\Phi_0\rangle$ we use the following adiabatic switch-on

$$\hat{H}(\tau) = \hat{H}_s(\tau) + f_\epsilon(t)H_1(\tau) \quad (26)$$

where $f_\epsilon = e^{\epsilon(t-t_0)}$ for $t < t_0$ and $f_\epsilon = 1$ for $t \geq t_0$. We further defined $\hat{H}_1(\tau) = \hat{H}(\tau) - \hat{H}_s(\tau)$. Note that in the interval $[-\infty, t_0]$ the Hamiltonian is the same on both sides of the contour. Using the Gell-Mann-Low theorem we then have

$$|\Psi_0\rangle = \lim_{\epsilon\to 0} A^{-1}V(\tau_i,-\infty)|\Phi_0\rangle = \lim_{\epsilon\to 0} A^{-1}V(\tau_f,+\infty)|\Phi_0\rangle \quad (27)$$

with the phase factor $A = \langle\Phi_0|V(\tau_i,-\infty)|\Phi_0\rangle = \langle\Phi_0|V(\tau_f,+\infty)|\Phi_0\rangle$. We then have (see Eq.(11)

$$\langle\mathbf{j}_p(\bar{1})\rangle = \lim_{\epsilon\to 0}\frac{\langle\Phi_0|V(+\infty,\tau)\mathbf{j}_p(\mathbf{r}_1)V(\tau,-\infty)|\Phi_0\rangle}{\langle\Phi_0|V(+\infty,-\infty)|\Phi_0\rangle} \quad (28)$$

where we used that $|A| = 1$. Expanding the evolution operator V in powers of $\hat{H}_1$ we find that

$$n(\bar{1})(\mathbf{A}_s(\bar{1}) - \mathbf{A}(\bar{1})) = \lim_{\epsilon\to 0}\sum_{n=1}^{\infty}\frac{(-i)^n}{n!}\int_C d\bar{2}\ldots\int_C d(\overline{n+1})$$
$$\times\, f_\epsilon(2)\ldots f_\epsilon(n+1)\langle T_C\Delta\mathbf{j}_p(\bar{1})\Delta\hat{H}_1(\bar{2})\ldots\Delta\hat{H}_1(\overline{n+1})\rangle_0 \qquad (29)$$

where the fluctuation operators $\Delta\hat{O} = \hat{O} - \langle\hat{O}\rangle_0$ appear from the expansion of the denominator in Eq.(28). The right hand side can now by Wick's theorem be expanded in Keldysh Green functions defined by

$$G(\bar{1},\bar{2}) = \langle\Phi_0|T_C\hat{\psi}(\bar{1})\hat{\psi}^\dagger(\bar{2})|\Phi_0\rangle$$
$$= \theta(\tau_1-\tau_2)G^>(1,2) + \theta(\tau_2-\tau_1)G^<(1,2) \qquad (30)$$

Eq.(29) is not directly usable, since we use a reference Hamiltonian that is determined self-consistently. This can however be resolved by an expansion of $\mathbf{A}_s$ in powers of the Coulomb interaction. If we denote the n-th order term by $\mathbf{A}_s^{(n)}$ we have (the Hartree part is explicitly a first order term) to first order:

$$n(\bar{1})(\mathbf{A}_H + \mathbf{A}_{xc}^{(1)}) = -i\lim_{\epsilon\to 0}\int_C d\bar{2}f_\epsilon(\bar{2})\langle T_C\Delta\mathbf{j}_p(\bar{1})\Delta\hat{W}(\bar{2})\rangle_0$$
$$+\, i\lim_{\epsilon\to 0}\int d\bar{2}f_\epsilon(\bar{2})\langle T_C\Delta\mathbf{j}_p(\bar{1})\Delta\mathbf{j}_p(\bar{2})\rangle_0\cdot(\mathbf{A}_H(\bar{2}) + \mathbf{A}_{xc}^{(1)}(\bar{2})) \qquad (31)$$

This can be written as

$$\lim_{\epsilon\to 0}\sum_\nu\int_C d\bar{2}f_\epsilon(\bar{2})\chi_{s,\mu\nu}(\bar{1},\bar{2})(A_{H,\nu}(\bar{2}) + A_{xc,\nu}^{(1)}(\bar{2})) =$$
$$-i\lim_{\epsilon\to 0}\int_C d\bar{2}f_\epsilon(\bar{2})\langle T_C\Delta\hat{\mathbf{j}}_{p,\mu}(\bar{1})\Delta\hat{W}(\bar{2})\rangle_0 \qquad (32)$$

If we evaluate this at physical potentials, then we can replace $\Delta\hat{O}$ by $\hat{O}$ (the average value is the same on both sides of the contour and integrates to zero). If we then use Wick's theorem and the explicit form of $\mathbf{A}_H$ we obtain

$$\lim_{\epsilon\to 0}\sum_\nu\int_C d\bar{2}f_\epsilon(\bar{2})\chi_{s,\mu\nu}(\bar{1},\bar{2})A_{xc,\nu}^{(1)}(\bar{2}) =$$
$$\lim_{\epsilon\to 0}2\int_C d\bar{2}\int_C d\bar{3}f_\epsilon(2)v(\bar{3},\bar{2})G(\bar{3},\bar{2})G(\bar{2},\bar{1})\mathbf{j}_{\mathbf{op},\mu}(\mathbf{r}_1)G(\bar{1},\bar{3}) \qquad (33)$$

where we defined the operator $\mathbf{j}_{\mathbf{op},\mu} = 1/2i(\overrightarrow{\partial}_\mu - \overleftarrow{\partial}_\mu)$ and where the Green functions are for the spin unpolarized case. The first order change $\mathbf{A}_{xc}^{(1)}$ is

only due to the exchange diagram. We therefore denote $\mathbf{A}_{xc}^{(1)} = \mathbf{A}_x$. Let us now write these equations in physical time:

$$\lim_{\epsilon\to 0}\sum_{\nu}\int_{-\infty}^{+\infty} dt_2 \int d^3r_2 f_\epsilon(t_2)\chi_{R,s,\mu\nu}(1,2)A_{x,\nu}(2) =$$
$$= \lim_{\epsilon\to 0} 2\int d^3r_2 \int d^3r_3 \int_{-\infty}^{t_1} dt f_\epsilon(t)\frac{G^<(\mathbf{r}_2 t,\mathbf{r}_3 t)}{|\mathbf{r}_2-\mathbf{r}_3|}$$
$$\times\left[G^<(\mathbf{r}_2 t,\mathbf{r}_1 t)\mathbf{j}_{\mathbf{op},\mu}(\mathbf{r}_1)G^>(\mathbf{r}_1 t,\mathbf{r}_3 t) - (>\leftrightarrow<)\right] \quad (34)$$

These equations are the extension of the time-dependent optimized potential method[14] (TDOPM) to current-density-functional theory. Note that for times $t \le t_0$ the time integrations can be done analytically as in this interval the time-dependence of the orbitals is known. After taking the limit $\epsilon \to 0$ we then obtain the static x-only OPM equations (which can be gauged to scalar form as there are no transverse external fields in the ground state). By collecting terms higher order in the two-particle interaction we can obtain equations for the higher order terms $A_{xc}^{(n)}$. On the other hand, we can by functional differentiation of Eq.(33) also obtain an integral equation for the kernel $f_{xc,\mu\nu}^{(1)}$. It would go to far to represent the details in the short presentation here. The function has recently been analyzed in detail for the electron gas [15] with the aim of providing an analytic form that can be used in the local density approximation for response calculations on inhomogeneous systems. We finally remark that the TDOPM equations can also be obtained from the so-called Keldysh Sham-Schlüter equation [16] in which case one can derive conserving approximations by means of Φ-derivable [17] and Ψ-derivable functionals [18].

References

1. E.K.U.Gross, J.F.Dobson and M.Petersilka, in *Topics in Current Chemistry.* (ed.) R.F.Nalewajski, Springer, (1996)
2. R.van Leeuwen, *Int.J.Mod.Phys.* **B15**, 1969 (2001).
3. G.Vignale and W.Kohn, *Phys.Rev.Lett.* **77**, 2037 (1996)
4. F.Kootstra, P.L.de Boeij and J.G.Snijders, *Phys.Rev.* **B62**, 7071 (2000)
5. C.A.Ullrich and G.Vignale, *Phys.Rev.***B65**, 245102 (2002)
6. M.van Faassen, P.L.de Boeij, R.van Leeuwen, J.A.Berger and J.G.Snijders, *Phys.Rev.Lett.* **88**, 186401 (2002)
7. L.I.Schiff, *Quantum Mechanics*, McGraw-Hill, (1968)
8. J.D.Bjorken and S.D.Drell, *Relativistic Quantum Fields*, McGraw-Hill, (1965)
9. J.Schwinger, *J.Math.Phys.* **2**, 407 (1961)
10. L.V.Keldysh, *Sov.Phys.JETP* **20**, 1018 (1965)
11. P.Danielewicz. *Ann.Phys.* **152**, 239, (1984)
12. J.Rammer and H.Smith. *Rev. Mod. Phys.*, **58**, 323 (1986)

13. R.van Leeuwen, *Phys. Rev. Lett.* **80**, 1280 (1998).
14. C. A. Ullrich, U. J. Gossmann, and E. K. U. Gross *Phys. Rev. Lett.* **74**, 872 (1995).
15. R.van Leeuwen, S.Kurth and U.von Barth, to be published
16. R.van Leeuwen, *Phys.Rev.Lett.* **76**, 3610 (1996).
17. G.Baym *Phys.Rev.* **127**, 1391 (1962)
18. C.-O. Almbladh, U.von Barth and R.van Leeuwen, *Int.J.Mod.Phys.***B13**, 535 (1999)